

RENEWAL PROPOSAL
AND
PROGRESS REPORT #2

ENTITLED

LABORATORY EVALUATION AND APPLICATION OF
MICROWAVE ABSORPTION PROPERTIES UNDER
SIMULATED CONDITIONS FOR PLANETARY ATMOSPHERES

to the

Planetary Atmospheres Program of the
National Aeronautics and Space Administration
for Grant NAG5-4190

Principal Investigator:
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Report Period: January 1, 1998 through December 31, 1998

Proposed Renewal Period: January 1, 1999 through December 31, 2001

Submitted: June 1998

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**PLANETARY ATMOSPHERES
COVER PAGE**
ROSS-98 NRA 98-OSS-03

Confirmation #: 99-053
Date Received: May 14, 1998

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Proposal Type: New Proposal

Grant/Contract/RTOP #: NAG5-4190

Proposal Category: Composition and Structure

Major Equipment Proposal? No

Do you intend to submit an E/PO Proposal? Yes

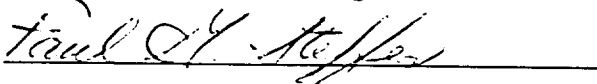
Proposal Title

Laboratory Evaluation and Application of Microwave Absorption
Properties under Simulated Conditions for Planetary Atmospheres

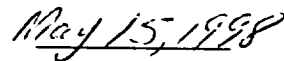
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Date



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Proposal Collaborator(s)

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E-mail Address

Institutional Endorsement

Certification of Compliance with Applicable Executive Orders and U.S. Code

By submitting the proposal identified in this *Cover Sheet/Proposal Summary* in response to NRA 98-OSS-03, the Authorizing Official of the proposing institution (or the individual proposer if there is no proposing institution) as identified below:

certifies that the statements made in this proposal are true and complete to the best of his/her knowledge;

agrees to accept the obligations to comply with NASA award terms and conditions if an award is made as a result of this proposal; and

confirms compliance with all provisions, rules, and stipulations set forth in the three Certifications contained in this NRA [namely, (i) *Certification Regarding Debarment, Suspension, and Other Responsibility Matters Primary Cover Transactions*, (ii) *Certification Regarding Lobbying*, and (iii) *Certification of Compliance with the NASA Regulations Pursuant to Nondiscrimination in Federally Assisted Programs*].

Willful provision of false information in this proposal and/or its supporting documents, or in reports required under an ensuing award, is a criminal offense (U.S. Code, Title 18, Section 1001).

Authorizing Official

Ms. Janis L. Goddard

Contracting Officer

Georgia Tech Research Corporation

Georgia Institute of Technology

Office of Contract Administration

Program Initiation Division

Atlanta, GA 30332-0420

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Type of Institution: Educational Institution

Signature

Date

Science Budget Summary

<i>Amount Requested:</i>	<i>Year 1999</i>	<i>Year 2000</i>	<i>Year 2001</i>	<i>Total Funding</i>
	\$92,417	\$92,417	\$92,417	\$277,251

E/PO Budget Summary

<i>Amount Requested:</i>	<i>Year 1999</i>	<i>Year 2000</i>	<i>Year 2001</i>	<i>Total Funding</i>
	\$10,000	\$10,000	\$10,000	\$30,000

PLANETARY ATMOSPHERES PROPOSAL SUMMARY

ROSS-98 NRA 98-OSS-03

Confirmation #: 99-053

Date Received: May 14, 1998

Proposal Summary

Radio absorptivity data for planetary atmospheres obtained from spacecraft radio occultation experiments, entry probe radio signal absorption measurements, and earth-based radio astronomical observations can be used to infer abundances of microwave absorbing constituents in those atmospheres, as long as reliable information regarding the microwave absorbing properties of potential constituents is available. The use of theoretically-derived microwave absorption properties for such atmospheric constituents, or using laboratory measurements of such properties taken under environmental conditions which are significantly different than those of the planetary atmosphere being studied, often leads to significant misinterpretation of available opacity data. For example, laboratory measurements completed recently by Kolodner and Steffes (ICARUS 132, pp.151-169, March 1998, attached as Appendix A) under this grant (NAG5-4190), have shown that the opacity from gaseous H₂SO₄ under simulated Venus conditions is best described by a different formalism than was previously used. The recognition of the need to make such laboratory measurements of simulated planetary atmospheres over a range of temperatures and pressures which correspond to the altitudes probed by both spacecraft entry probe and orbiter radio occultation experiments and by radio astronomical observations, and over a range of frequencies which correspond to those used in such experiments, has led to the development of a facility at Georgia Tech which is capable of making such measurements. It has been the goal of this investigation to conduct such measurements AND to apply the results to a wide range of planetary observations, both spacecraft and earth-based, in order to determine the identity and abundance profiles of constituents in those planetary atmospheres.

Recent accomplishments and key activities over the new three-year grant cycle include:

A. Venus Studies

1. Completion of laboratory measurement programs of microwave absorption properties of SO₂ and gaseous H₂SO₄: New formalisms for their opacity, with application to radio observations were published in Suleiman et al. (JGR-Planets 101, pp. 4623-4635, Feb 1996) and in Kolodner and Steffes (ICARUS 132, pp.151-169, March 1998). 2. Maps of the microwave emission of Venus were made in April 1996, and in December 1997 (jointly with NIR observers). Radiative transfer models have been developed which show that the substantial polar darkening corresponds to elevated abundances of gaseous H₂SO₄. This is consistent with results from Magellan radio occultation experiments, conducted with partial support from this grant. 3. A key task for the first year of the next grant cycle is completion of radiative transfer analysis of the VLA maps, and intercomparison with NIR images and radio occultation results from Magellan. This analysis will give important insight into constituent abundance variations, and to dynamical models for the Venus atmosphere.

B. Outer Planets Studies:

1. A radiative transfer model was developed for the atmosphere of Neptune and indicates the possible role of phosphine (PH₃) in explaining opacity measured in that atmosphere (DeBoer and Steffes, ICARUS 123, pp. 324-335, October 1996). 2. Our laboratory measurement system has been substantially renovated and upgraded, so as to allow high accuracy measurement of the microwave absorptivity and refractivity of phosphine under simulated conditions for the outer planets. 3. In the first two years of the next grant cycle, we will

conduct measurements of the microwave opacity and refraction of phosphine under simulated conditions for the outer planets. We will then develop a formalism describing this opacity as a function of frequency, temperature, and pressure, and then apply the results to absorptivity data from the Galileo Probe (deep atmosphere of Jupiter), to our radiative transfer mode. for interpreting the microwave emission from the outer planets, to older Voyager radio occultation data, and to aid in the planning of the Cassini Saturn encounter. 4. In the third year of the next grant cycle, we plan to measure the millimeter-wavelength opacity of phosphine, to be used in planning possible effects on the Ka-Band radio scientific experiments to be conducted by Cassini at Saturn, and in the interpretation of new millimeter-wavelength emission maps of the outer planets to be obtained from new array telescopes.

SUMMARY OF PERSONNEL COMMITMENTS AND COSTS

	<u>1999</u>	<u>2000</u>	<u>2001</u>
Paul G. Steffes, Ph.D.			
PRINCIPAL INVESTIGATOR -- COST	\$29K	\$29K	\$29K
(Man Year effort)	(0.25)	(0.25)	(0.25)
James P. Hoffman			
GRADUATE RESEARCH ASSISTANT--COST	\$18K	\$18K	\$18K
(Man Year effort)	(0.5)	(0.5)	(0.5)

Principal Investigator: Paul G. Steffes (Georgia Institute of Technology)
 Title: Laboratory Evaluation and Application of Microwave Absorption Properties under Simulated
 Conditions for Planetary Atmospheres
 Grant Number: NAG5-4190

BUDGET SUMMARY

For the period from January 1, 1999 to December 31, 1999

		<u>NASA USE ONLY</u>		
		A	B	C
1.	<u>Direct Labor*</u>	<u>\$ 57,483</u>	_____	_____
A.	Principal Investigator (Paul G. Steffes) 25% time, calendar year (.25 person-years) \$ 28,525			
B.	1 Graduate Student (J. P. Hoffman) 50% time, calendar year (.5 person-years) \$ 17,856			
C.	1 Senior Administrative Secretary 10% time, calendar year (.10 person-years) \$ 3,000			
D.	Fringe benefits (25.7% of direct salaries and wages, less students) \$ 8,102			
2.	<u>Other Direct Costs:</u>			
a.	Supplies (Gas mixtures, microwave connectors, o-rings and data storage media for lab experiments) <u>\$ 1,500</u>		_____	_____
b.	Travel (For graduate student and principal investigator: Attend 1999 AAS/DPS Meeting --5 days, Padova, Italy. Airfare: \$750 plus registration and \$100 /day for food and lodging) <u>\$ 3,000</u>		_____	_____
3.	<u>Facilities and Administrative Costs:</u>	<u>\$30,434</u>	_____	_____
	Overhead (indirect expense), 49.1% of direct cost base **			
4.	<u>Other Applicable Costs:</u>	<u>\$ 3,296</u>	_____	_____
	Tuition Remission (\$824 per student per quarter)			
5.	<u>SUBTOTAL -- Estimated Costs</u>	<u>\$92,417</u>	_____	_____
6.	<u>Education/Public Outreach (E/PO) Activity:</u>	<u>\$10,000</u>	_____	_____
	(See enclosed E/PO Proposal for budget details)			
7.	<u>Total Estimated Costs: ***</u>	<u>\$102,417</u>	_____	XXXX
8.	APPROVED BUDGET	XXXXXX	XXXX	_____

* The salary and wage rates are based on FY99 salaries for the Georgia Institute of Technology. The Georgia Tech Fiscal Year is July 1 through June 30.

**Rates are for the period July 1, 1998 through June 30, 1999, and are subject to adjustment upon DCAA audit and ONR negotiations.

***No cost sharing or carryover funds are proposed for this program.

Principal Investigator: Paul G. Steffes (Georgia Institute of Technology)
 Title: Laboratory Evaluation and Application of Microwave Absorption Properties under Simulated
 Conditions for Planetary Atmospheres
 Grant Number: NAG5-4190

BUDGET SUMMARY
For the period from January 1, 2000 to December 31, 2000

		<u>NASA USE ONLY</u>		
		A	B	C
1. <u>Direct Labor*</u>		<u>\$ 57,483</u>	_____	_____
A.	Principal Investigator (Paul G. Steffes) 25% time, calendar year (.25 person-years)	\$ 28,525		
B.	1 Graduate Student (J. P. Hoffman) 50% time, calendar year (.5 person-years)	\$ 17,856		
C.	1 Senior Administrative Secretary 10% time, calendar year (.10 person-years)	\$ 3,000		
D.	Fringe benefits (25.7% of direct salaries and wages, less students)	\$ 8,102		
2. <u>Other Direct Costs:</u>				
a.	Supplies (Gas mixtures, microwave connectors, o-rings and data storage media for lab experiments)	<u>\$ 1,500</u>	_____	_____
b.	Travel (For graduate student and principal investigator: Attend AAS/DPS Meeting --5 days, Pasadena, CA. Airfare: \$750 plus registration and \$100 /day for food and lodging)	<u>\$ 3,000</u>	_____	_____
3. <u>Facilities and Administrative Costs:</u>				
	Overhead (indirect expense), 49.1% of direct cost base **	<u>\$30,434</u>	_____	_____
4. <u>Other Applicable Costs:</u>				
	Tuition Remission (\$824 per student per quarter)	<u>\$ 3,296</u>	_____	_____
5. <u>SUBTOTAL -- Estimated Costs</u>		<u>\$92,417</u>	_____	_____
6. <u>Education/Public Outreach (E/PO) Activity:</u>				
	(See enclosed E/PO Proposal for budget details)	<u>\$10,000</u>	_____	_____
7. <u>Total Estimated Costs: ***</u>		<u>\$102,417</u>	_____	XXXX
8. <u>APPROVED BUDGET</u>		XXXXXX	XXXX	_____

* The salary and wage rates are based on FY99 salaries for the Georgia Institute of Technology. The Georgia Tech Fiscal Year is July 1 through June 30.

**Rates are for the period July 1, 1998 through June 30, 1999, and are subject to adjustment upon DCAA audit and ONR negotiations.

***No cost sharing or carryover funds are proposed for this program.

Principal Investigator: Paul G. Steffes (Georgia Institute of Technology)
 Title: Laboratory Evaluation and Application of Microwave Absorption Properties under Simulated
 Conditions for Planetary Atmospheres
 Grant Number: NAG5-4190

BUDGET SUMMARY

For the period from January 1, 2001 to December 31, 2001

		<u>NASA USE ONLY</u>		
		A	B	C
1.	<u>Direct Labor*</u>	<u>\$ 57,483</u>	_____	_____
A.	Principal Investigator (Paul G. Steffes) 25% time, calendar year (.25 person-years) \$ 28,525			
B.	1 Graduate Student (J. P. Hoffman) 50% time, calendar year (.5 person-years) \$ 17,856			
C.	1 Senior Administrative Secretary 10% time, calendar year (.10 person-years) \$ 3,000			
D.	Fringe benefits (25.7% of direct salaries and wages, less students) \$ 8,102			
2.	<u>Other Direct Costs:</u>			
a.	Supplies (Gas mixtures, microwave connectors, o-rings and data storage media for lab experiments) <u>\$ 1,500</u>		_____	_____
b.	Travel (For graduate student and principal investigator: Attend AAS/DPS Meeting --5 days, Pasadena, CA. Airfare: \$750 plus registration and \$100 /day for food and lodging) <u>\$ 3,000</u>		_____	_____
3.	<u>Facilities and Administrative Costs:</u>	<u>\$30,434</u>	_____	_____
	Overhead (indirect expense), 49.1% of direct cost base **			
4.	<u>Other Applicable Costs:</u>	<u>\$ 3,296</u>	_____	_____
	Tuition Remission (\$824 per student per quarter)			
5.	<u>SUBTOTAL -- Estimated Costs</u>	<u>\$92,417</u>	_____	_____
6.	<u>Education/Public Outreach (E/PO) Activity:</u>	<u>\$10,000</u>	_____	_____
	(See enclosed E/PO Proposal for budget details)			
7.	<u>Total Estimated Costs: ***</u>	<u>\$102,417</u>	_____	XXXX
8.	APPROVED BUDGET	XXXXXX	XXXX	_____

* The salary and wage rates are based on FY99 salaries for the Georgia Institute of Technology. The Georgia Tech Fiscal Year is July 1 through June 30.

**Rates are for the period July 1, 1998 through June 30, 1999, and are subject to adjustment upon DCAA audit and ONR negotiations.

***No cost sharing or carryover funds are proposed for this program.

Principal Investigator: Paul G. Steffes

Current Grant No: NAG5-4190 (This proposal serves also as Progress Report #2 for Grant NAG5-4190)

Title: Laboratory Evaluation and Application of Microwave Absorption Properties under Simulated Conditions for Planetary Atmospheres

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I. INTRODUCTION AND SUMMARY

Radio absorptivity data for planetary atmospheres obtained from spacecraft radio occultation experiments, entry probe radio signal absorption measurements, and earth-based radio astronomical observations can be used to infer abundances of microwave absorbing constituents in those atmospheres, as long as reliable information regarding the microwave absorbing properties of potential constituents is available. The use of theoretically-derived microwave absorption properties for such atmospheric constituents, or using laboratory measurements of such properties taken under environmental conditions which are significantly different than those of the planetary atmosphere being studied, often leads to significant misinterpretation of available opacity data. For example, laboratory measurements completed recently by Kolodner and Steffes (1998, reprint attached as Appendix A) under this grant (NAG5-4190), have shown that the opacity from gaseous H_2SO_4 under simulated Venus conditions is best described by a different formalism than was previously used. The recognition of the need to make such laboratory measurements of simulated planetary atmospheres over a range of temperatures and pressures which correspond to the altitudes probed by both radio occultation experiments and radio astronomical observations, and over a range of frequencies which correspond to those used in both spacecraft entry probe and orbiter (or flyby) radio occultation experiments and radio astronomical observations, has led to the development of a facility at Georgia Tech which is capable of making such measurements. It has been the goal of this investigation to conduct such measurements *and* to apply the results to a wide range of planetary observations, both spacecraft and earth-based, in order to determine the identity and abundance profiles of constituents in those planetary atmospheres.

II. VENUS STUDIES

A. Laboratory Measurements under Simulated Venus Conditions

From 1991 through 1994, as part of this project, we were active in using the Magellan spacecraft to probe the Venus atmosphere by way of radio occultation studies (Steffes *et al.*, 1994). One key aspect of the Magellan radio occultation results is the high percentage accuracy of the measured profiles of 13 cm and 3.6 cm absorptivity; typically ± 10 -15% (Jenkins *et al.*, 1994). To take advantage of these new profiles, so as to develop highly accurate abundance profiles of the microwave absorbing constituents, one must know the microwave absorbing and refracting properties of the constituents very accurately. At 13 cm, the opacity immediately below the clouds is almost all due to gaseous sulfuric acid (H_2SO_4). (See, e.g., Steffes, 1985.) Sulfuric acid is, of course, the predominant constituent in the Venus clouds. Understanding the spatial and temporal variations in its gas-phase abundance gives insight into the dynamical processes which affect cloud formation, as well as into the thermochemical processes which constrain the abundances of other reactive constituents in the Venus atmosphere such as COS, H_2O , CO, SO_2 , and SO_3 . Over the past grant year, we completed our analysis and published

our newest laboratory measurements of the absorption and refraction of gaseous H_2SO_4 in a CO_2 atmosphere under simulated Venus conditions, at selected wavelengths from 1.3 to 13.5 cm (frequencies from 2.2 to 22 GHz). The results of these laboratory measurements, with their application to the interpretation of Magellan and Mariner 10 radio occultation microwave absorptivity profiles, yielding abundance profiles for gaseous H_2SO_4 , are given in a paper by Kolodner and Steffes (1998), which appeared in the March 1998 issue of *ICARUS*, and is attached as Appendix A. It should be noted that in order to obtain the high accuracies required to accurately interpret the radio occultation results, substantial upgrades to our laboratory measurement system were made. Since some of these modifications actually reduced the sensitivity of the system (in order to prevent reactions of the hot sulfuric acid gas with metals inside the microwave measurement system), they are now in process of being changed, so as to optimize our system for upcoming measurements of simulated outer planets atmospheres. (The laboratory system is described in Section III.B and in the appended papers.) These laboratory results are being used by our group, and other investigators, in the interpretation of the additional radio occultation measurements taken using the Magellan spacecraft in Summer 1994 (Jenkins, NASA-Ames/SETI Institute, and Steffes, Georgia Tech), and in the interpretation of NRAO/VLA microwave emission maps of Venus taken in April 1996 and December 1997. (See Section II.B below).

B. Venus Observations and Radiative Transfer Modelling

In October 1995, a proposal was submitted to the National Radio Astronomy Observatory (NRAO) for use of the Very Large Array (VLA) for mapping the 1.3 cm and 2 cm emission from Venus. These wavelengths were chosen since they are especially sensitive to the opacity from SO_2 and H_2SO_4 . (Ref: Suleiman *et al.*, 1996 and Kolodner and Steffes, 1998, appended.) The observations were conducted on April 5, 1996 by graduate students Shady H. Suleiman and Marc A. Kolodner, assisted by Dr. Brian Butler from NRAO. In order to process the data taken with the 27 elements of the VLA into brightness maps, a relatively standard procedure was used whereby cross correlations from individual pairs of antennas were measured over the 12 hour observing period. These quantities, known as "visibilities" are then mapped onto a two dimensional plane, known as the antenna coordinates plane, or u-v plane, and are then calibrated relative to the flux of spatially compact reference stars. (In this case, a nearby radio source, 0403+260 was observed every 5.5 minutes to calibrate out short-term variabilities in the array. Absolute calibration scales were obtained by referencing this "secondary" calibrator to "primary" calibrators 3C286 and 3C48.) By taking the two-dimensional Fourier transform of the visibilities, or coherence function, spatial maps of the planetary brightness are then obtained. In our case the maps of brightness at 2 cm (U-Band) and 1.3 cm (K-Band) show relatively uniform disks, with disk averaged emissions of 565.8 ± 17 K and 499.1 ± 25 K, respectively at 2 cm and 1.3 cm. Note that the 3-5% uncertainties in the disk-averaged flux are due largely to the uncertainty in the absolute flux of the calibration sources. The actual "noise" on the images is only on the order of 1 K, or 0.2%. The spatial resolution of the VLA synthesized beams are 1.45 arcseconds at 2 cm (U-

Band) and 1.1 arcseconds at 1.35 cm (K-Band). These maps of the continuum emission are shown in Figure 1.

Of more interest for the study of the Venus atmosphere however, is the detailed variation of the microwave emission from various regions on the Venus disk. In order to study this in detail, two radiative transfer models were independently developed by two graduate students who recently completed their Ph.D.'s under this program (Suleiman, 1997 and Kolodner, 1997). In Figures 2 and 3 we show maps of the residual 2 cm and 1.35 cm emission from Venus. These residuals were obtained by subtracting out the emission from the Venus surface and the emission and absorption effects from CO₂, the dominant atmospheric constituent. These maps show additional limb darkening (relative to the center of the disk, or "sub-earth point") from constituents other than CO₂ on the order of 14 K at both wavelengths in the equatorial zones, but up to about 25 K at both wavelengths in the polar zones. This, of course implies a higher density of microwave absorbing constituents at high latitudes, or a substantial reduction in the temperature in the polar zones.

Fortunately, radio occultation measurements of both temperature structure and microwave absorptivity in high latitude regions were conducted as part of the Magellan radio occultation experiments (Steffes *et al.*, 1994; Jenkins *et al.*, 1994). By using abundances of gaseous H₂SO₄ derived from this data (see Kolodner and Steffes, 1998, attached as Appendix A), our radiative transfer model was used to compute expected limb darkening relative to a sub-earth point whose gas abundances are derived from an equatorial radio occultation experiment (i.e. Mariner 10, see Kolodner and Steffes, 1998, appended). Preliminary results from our radiative transfer model show that the limb darkening in the equatorial zones (i.e., 14 K) is consistent with the gaseous H₂SO₄ abundance derived from the Mariner 10 radio occultation. However, to explain the extra darkening in the polar zones, a larger and more vertically spread abundance of gaseous H₂SO₄, such as that derived from the Magellan radio occultation from orbit #3212 (67 degrees North latitude) is required. The consistency between the 1.3 and 2 cm radio emission maps from the VLA and radio occultation results at 3.6 and 13 cm is due to our better understanding of the microwave properties of gaseous H₂SO₄ and provides further verification of the latitudinally dependent abundance of gaseous H₂SO₄ inferred by Jenkins and Steffes (1990) from earlier Pioneer-Venus radio occultation measurements.

These preliminary results were presented at the August 1997 AAS/DPS Meeting (Kolodner *et al.*, 1997), and are being prepared for publication in *ICARUS* later this year.

C. Future Work

Upon presentation of the above work at the August 1997 AAS/DPS meeting, quite a bit of interesting discussion began with the groups involved with the near-IR imaging of the Venus atmosphere. (Meadows and Crisp from JPL and Hillman and Chanover from NASA/GSFC). While the darkened polar regions in our preliminary maps seemed consistent with the polar darkening observed at longer (thermal) IR wavelengths by the

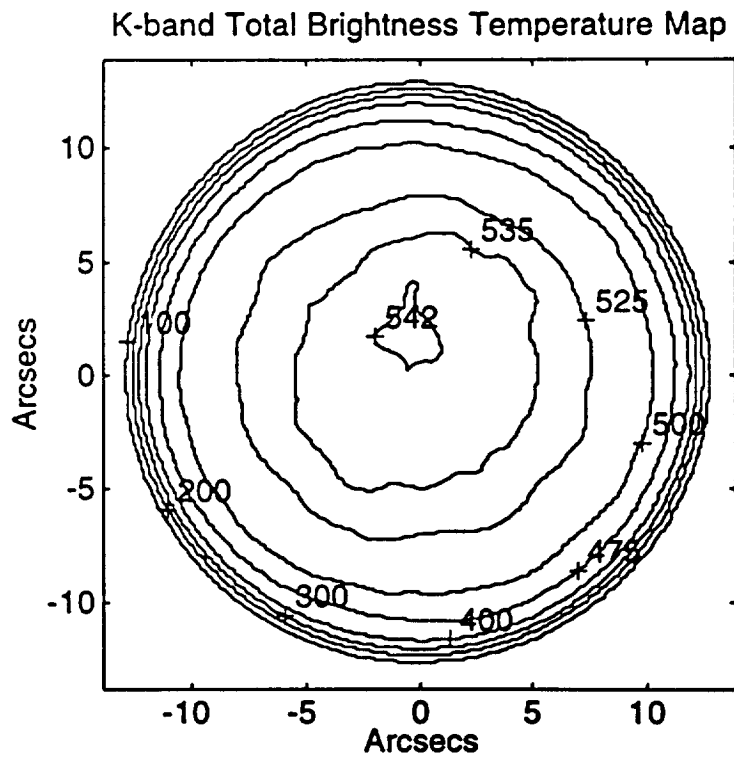
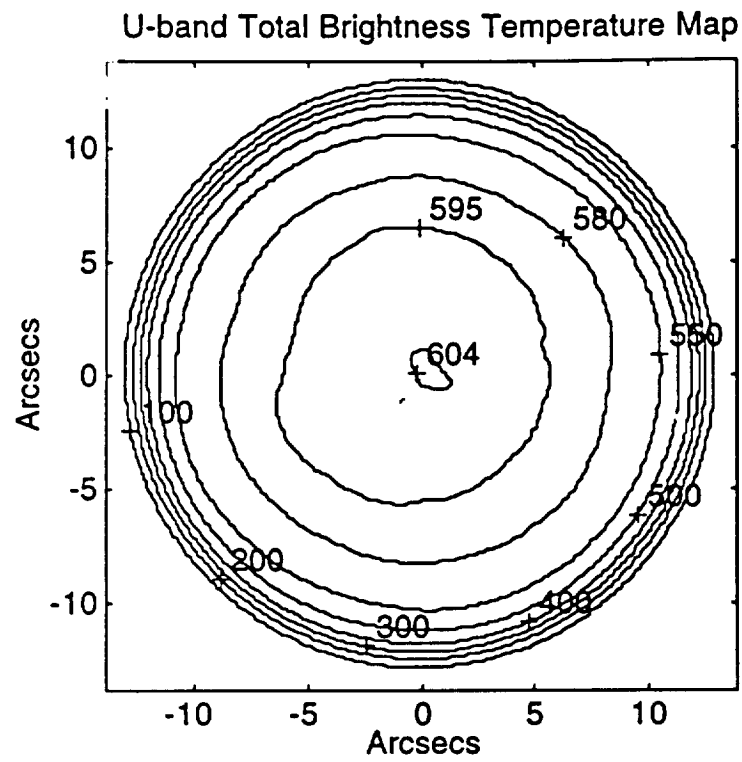


Figure 1: Contour plots of the total brightness temperature maps of Venus. The contour labels are given in units of Kelvins.

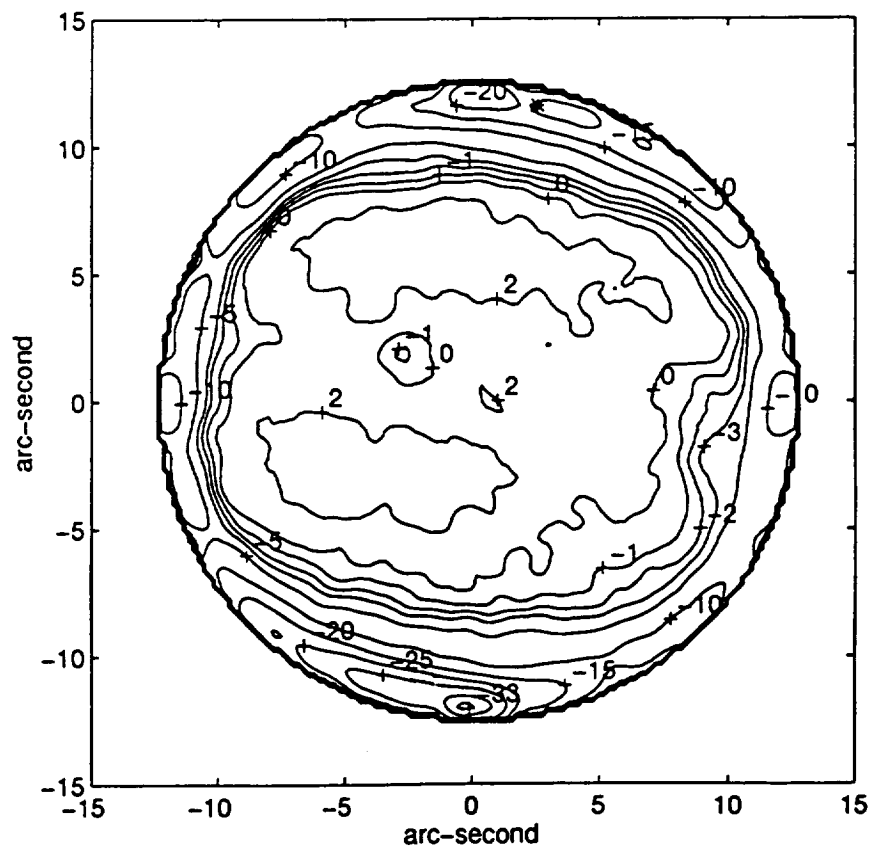
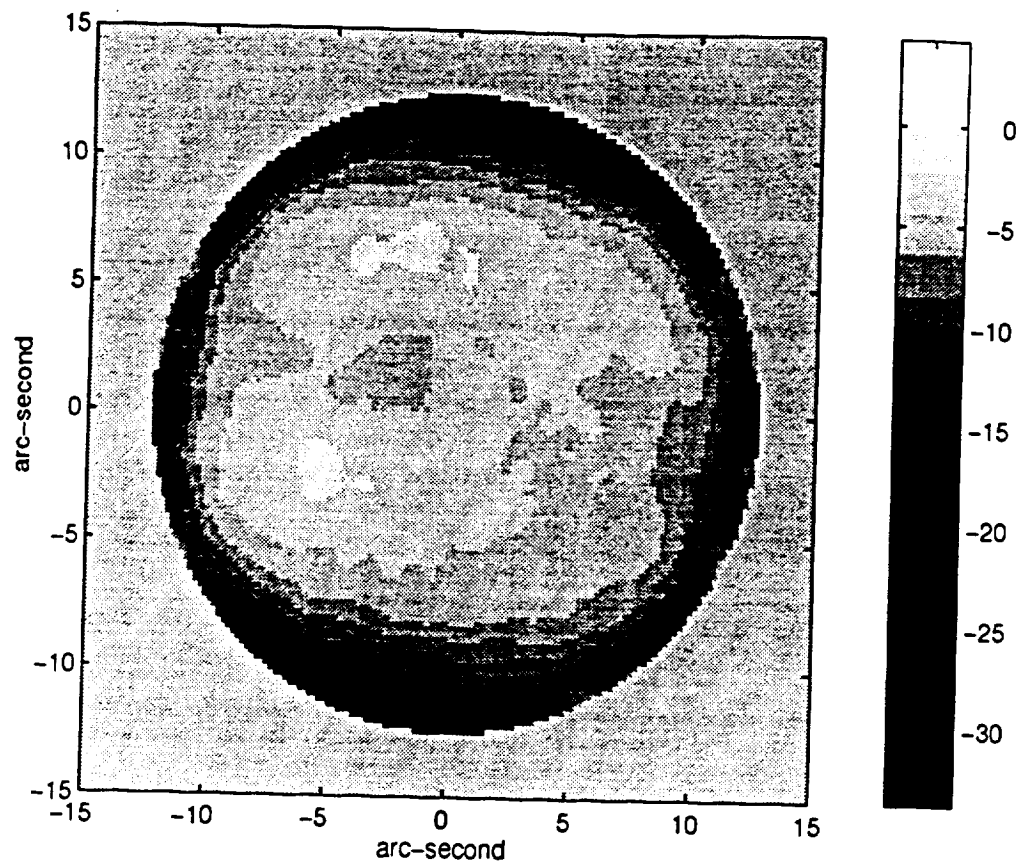


Figure 2 : Residual brightness temperature contour map of Venus at 14.94 GHz.

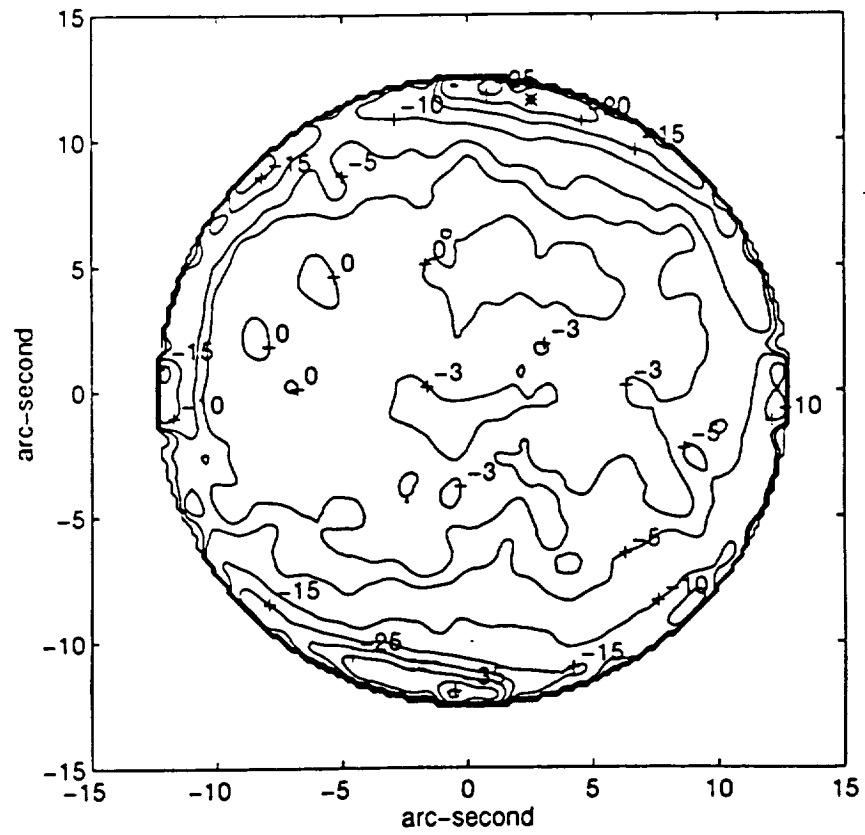
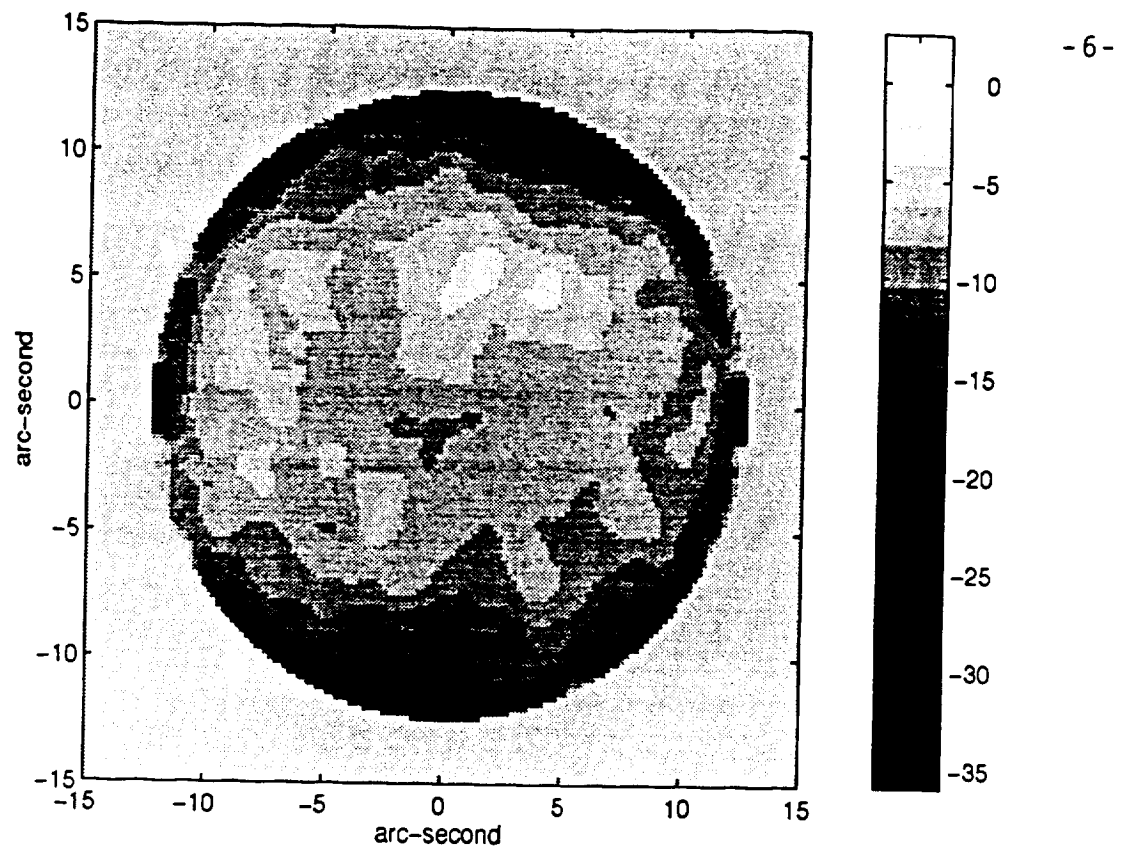


Figure 3 : Residual brightness temperature contour map of Venus at 22.46 GHz.

Pioneer Venus Orbiter Infrared Radiometer (OIR) experiment (Taylor, *et al.*, 1980), they appeared almost anti-correlated with Earth-based near-IR images (e.g., Hillman, *et al.*, 1996) whose features are more related to variations in integrated cloud density. However, it appeared that the combined results of microwave and IR mapping, along with radio occultation measurements, was consistent with a meridional Hadley cell circulation, which could potentially explain the latitudinal variations in the vertical distribution and abundance of gaseous H_2SO_4 measured by Mariner, Pioneer-Venus, and Magellan radio occultation experiments. Since the intercomparison of the IR and microwave maps was difficult, in that they were measured at different epochs, a proposal was submitted in September 1997 to the NRAO for use of the VLA for imaging of Venus simultaneously with near-IR observations conducted by Meadows and Crisp at ATNF (Australia) and by Hillman and Chanover at the Apache Point Observatory (New Mexico). (The proposal for this observation, including scientific justification, is attached as Appendix B.) Our measurement was conducted on December 20, 1997, jointly with Dr. Jon M. Jenkins (SETI Institute/ NASA Ames Research Center) and Dr. Bryan J. Butler (NRAO/VLA).

In the first grant year (1/1/99-12/31/99) of the new grant cycle, we will continue our interpretative studies of the Venus microwave radio emission maps and radio occultation studies, working with the other investigator groups (both IR and microwave), so as to yield the best possible understanding of the variations in constituent abundances and the atmospheric dynamics on a global basis. Now that our laboratory-based formalisms for the opacity of SO_2 (Suleiman *et al.*, 1996) and gaseous H_2SO_4 (Kolodner and Steffes, 1998) under Venus atmospheric conditions have been derived, new interpretative studies of both Venus radio occultation absorptivity profiles and Venus microwave radio emission maps have begun. The first paper describing these interpretive studies will be submitted to *ICARUS* later this year. We also expect to complete a higher-level dynamic model of the abundance and circulation of cloud forming gases when the results from the joint NIR and microwave mapping activities of December 1997 are complete. These results will be especially useful to the numerous potential Venus missions which are currently being proposed under the Discovery program, since many will involve NIR and microwave studies of the Venus atmosphere as part of their missions.

III. Outer Planets Studies

A. Phosphine in the Atmospheres of the Outer Planets

In our most recent study of the microwave emission spectrum of Neptune (DeBoer and Steffes, 1996a), we showed that in order to best match the most reliable disk-averaged emission measurements in the 1 mm to 20 cm range, and not exceed the measurements of 13 cm and 3.6 cm absorptivity made by Voyager 2 at Neptune (Lindal, 1992), a Neptune atmosphere where the abundance of H_2S is greater than that of NH_3 below the putative NH_4SH cloud in the deep atmosphere is required. While such an atmosphere (e.g. 78%

H₂, 19% He, 3% CH₄, plus 40 x solar H₂S and 0.2 x solar NH₃) gives an excellent fit to the microwave emission spectrum, its opacity is too low at 13 cm and 3.6 cm to explain the Voyager radio occultation results. It is possible, however, to match both emission spectra and the Voyager results by adding phosphine (PH₃) to the model.

An even more compelling problem is the large radio opacity detected in Jupiter's deep atmosphere by the radio transmissions from the Galileo entry probe (Folkner *et al.*, 1998). Based on the estimates of 21-cm wavelength opacity derived from the Galileo entry probe signal level as measured at the Galileo orbiter, Folkner *et al.* (1998) concluded that an abundance of ammonia four times the solar abundance is required to explain the measured opacity at pressures greater than 7 Bars, which is significantly larger than the deep atmospheric ammonia abundances inferred from IR observations and the vast majority of atmospheric models. While the Galileo team did consider possible contributions from H₂S and H₂O to the microwave opacity, they did not consider the effects of PH₃, largely because so little is known about its centimeter-wavelength opacity. Unfortunately, it was very difficult to detect PH₃ with the Galileo Probe mass spectrometer, since phosphine's molecular mass is identical to that of another constituent, H₂S.

Phosphine *has* been detected high in the atmospheres of Jupiter and Saturn from the millimeter-wavelength spectra at phosphine's strong rotational resonance (267 GHz, see Weisstein and Serabyn, 1994, 1996). Preliminary estimates with our Neptune emission model suggest that a PH₃ abundance between 10x and 20x solar best fits the centimeter wavelength microwave emission data. The amount of PH₃ required to explain the excess 21-cm opacity measured by the Galileo probe deep in Jupiter's atmosphere is difficult to estimate, since no lineshape data currently exists under those pressure conditions. For comparison, note that Weisstein and Serabyn (1994) inferred a 20x solar abundance at Saturn. Our estimates of the microwave absorption spectrum from PH₃ have been made using the updated Poynter, Pickett, and Cohen line catalog (1994). While some line intensities have been measured, many, including the weak inversion lines in the centimeter wavelength range, have not; and only one room-temperature measurement of line broadening parameters of one rotational resonance has been made (Pickett *et al.*, 1981). However, by assuming the Van Vleck-Weisskopf lineshapes and the Pickett *et al.* (1981) broadening parameters, it is possible to coarsely estimate the expected opacity.

In Figure 4, we show the expected opacity from an H₂/He/PH₃ mixture with an 8% abundance of phosphine under various temperature and pressure conditions representative of the outer planets. This figure shows that the centimeter wavelength opacity from the mixture including PH₃ will exceed the opacity from similar H₂S mixtures we successfully measured previously (DeBoer and Steffes, 1994). However, the actual opacity may vary by an order of magnitude depending on which lineshape parameters are used. Thus to accurately infer the PH₃ abundance in the atmospheres of the outer planets from centimeter-wavelength microwave data (e.g. radio telescopic observations), from spacecraft radio occultation experiments (e.g. Cassini at Saturn), and from entry probe

a. 0-1000 GHz

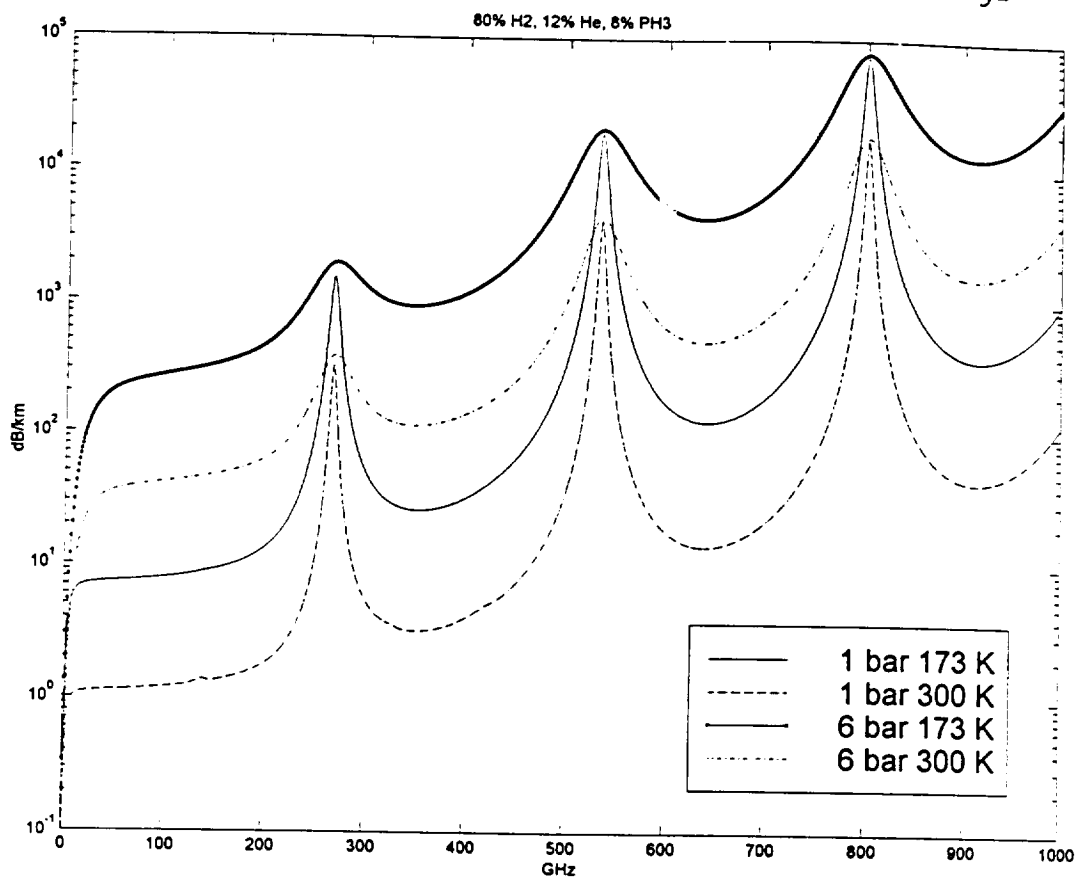
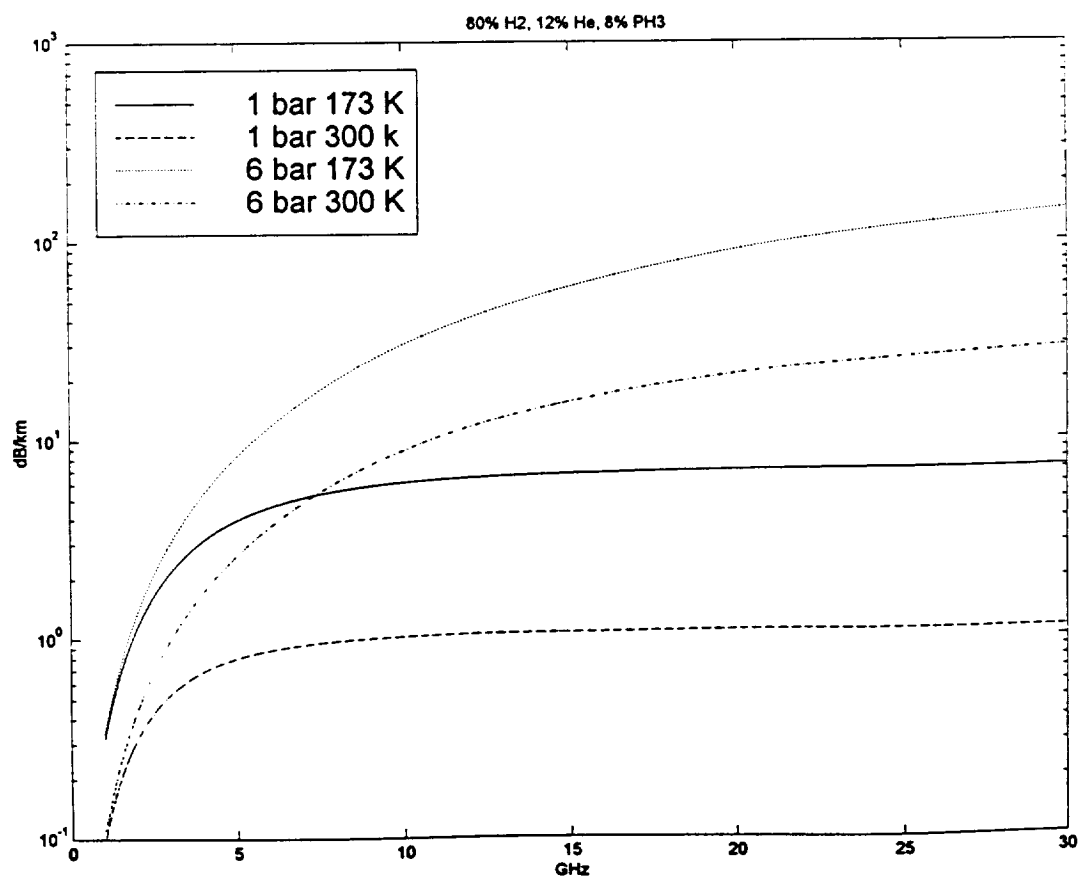


Figure 4: Predicted absorption from phosphine(PH₃ -- 8%) in an H₂/He atmosphere.

b. 0-30 GHz



uplink radio absorption data (e.g., Galileo at Jupiter) accurate laboratory measurements of phosphine's microwave opacity (and refractivity) are necessary.

B. Proposed Laboratory Measurements

Over the past grant year (January 1, 1998 thru December 31, 1998), we have renovated the system used previously by DeBoer and Steffes (1994) to measure the opacity and refractivity of H_2S under simulated conditions for the outer planets, and by Kolodner and Steffes (1998) to measure the opacity and refractivity of gaseous H_2SO_4 under simulated conditions for the Venus atmosphere, so as to measure the opacity and refractivity of PH_3 under simulated conditions for the outer planets. The renovations have included re-machining and re-plating the microwave resonators, as well as developing a PH_3 compatible gas handling system. A diagram of the renovated measurement system is shown in Figure 5. In order to further increase sensitivity and frequency range over our previous system (see DeBoer and Steffes, 1996b, attached as Appendix C), the Georgia Tech Research Corporation recently provided over \$90,000 to procure a new HP 8564E microwave spectrum analyzer, and a new HP 83650B synthesized microwave sweep generator. These new microwave components are currently being integrated into the system. The new system should be able to detect accurately the opacity from the mixture indicated in Figure 4, even if the opacity is one order of magnitude less than the nominal prediction.

As discussed in DeBoer and Steffes (1996b, attached as Appendix C), the refraction and absorptivity of a gas mixture at a particular wavelength can be determined by measuring the center frequency and bandwidth of a specific resonance of the cavity resonator with the gas mixture present, and then comparing it with the bandwidth and center frequency of the same resonance when a lossless reference gas is placed in the system. The measurement is repeated several times and averaged for each resonant frequency being tested, under the different temperature and pressure selected. Nitrogen is usually selected as the reference gas because of its very low microwave absorption and relatively large refractivity.

In the first year of the new grant cycle (January 1, 1999-December 1, 1999) The opacity and refractivity of a custom-manufactured (Matheson) gaseous mixture consisting of 82.8% H_2 , 9.2% He, and 8% PH_3 will be initially measured at five frequencies: 1.5 GHz (20 cm), 2.25 GHz (13.3 cm), 8.5 GHz (3.7 cm), 12.0 GHz (2.5 cm), and 21.7 GHz (1.38 cm) at pressures from 1 to 6 Bars. The first frequency (1.5 GHz) is close to the Galileo Probe transmitter frequency, and will be of direct use in interpreting that opacity. The next two, 2.25 GHz and 8.5 GHz correspond to the S-Band and X-Band radio transmission frequencies from both the Voyager and Cassini spacecraft, and thus will be of direct use in the interpretation of absorption measured by radio occultation experiments at all four Jovian planets. The two highest frequencies are close to the observing frequencies of the NRAO/VLA (Very Large Array), which has been used to image all of the giant planets. Thus, more accurate interpretation of those maps (using radiative transfer models such as ours) will be facilitated. The first year's measurements

Experimental Setup for Microwave Absorption in Outer Planet Conditions

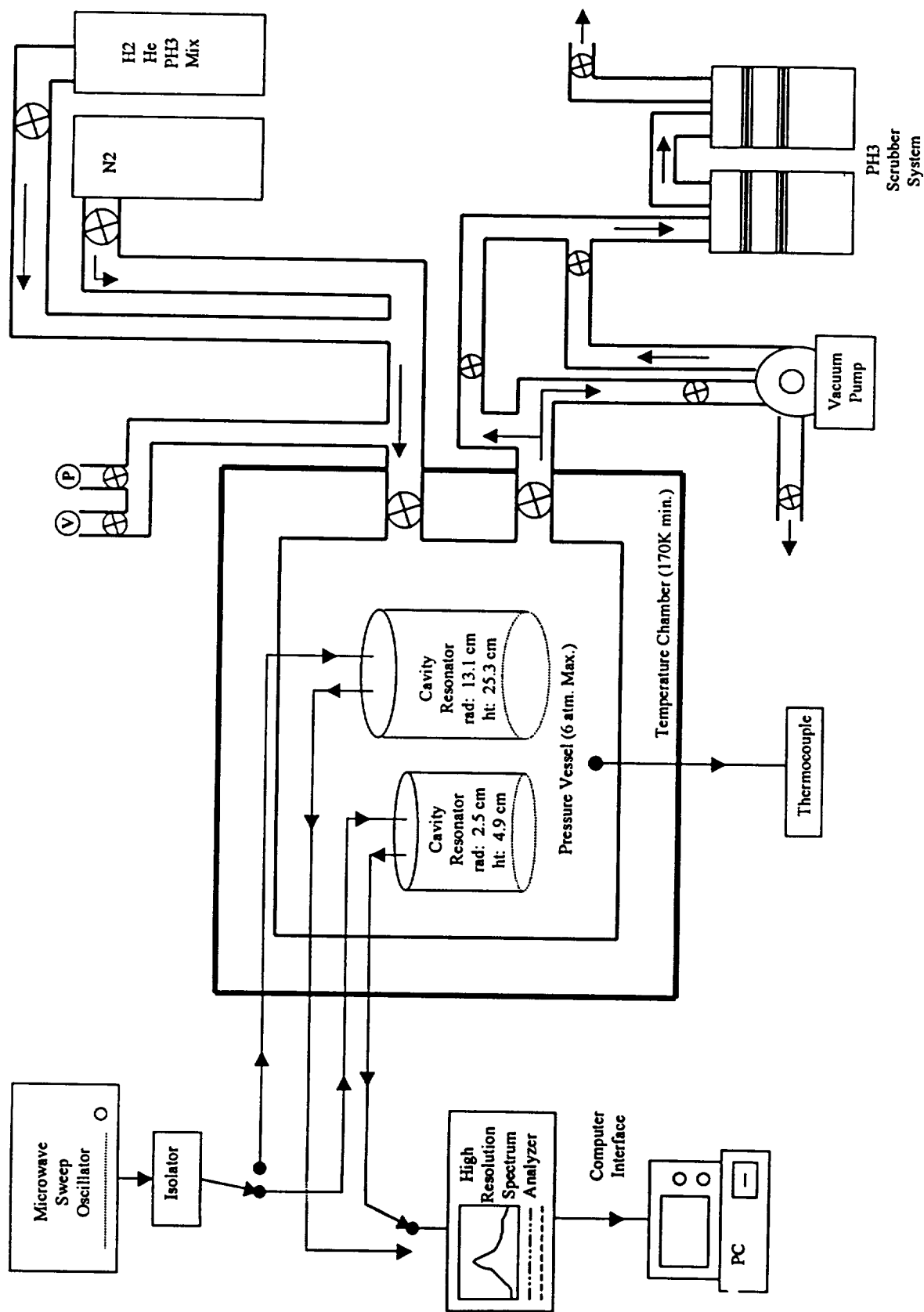


Figure 5

will be conducted at room temperature, both in order to better test the system, and because 300 K corresponds closely to the temperature in the deep atmosphere of Jupiter at which the Galileo probe detected large 21 cm opacity. In the second year of the project (January 1, 2000 thru December 31, 2000) measurements at 170 K and 210 K will be conducted at the above five wavelengths and over pressures from 1 to 6 Bars. The results will be analyzed, and will then be used to develop a formalism for PH_3 opacity similar to those we previously derived for ammonia, hydrogen sulfide, sulfur dioxide, and gaseous sulfuric acid. In the third year of the project (January 1, 2001 thru December 31, 2001), we will conduct measurements of the *millimeter* wavelength refraction and absorption from PH_3 , using the Fabry-Perot resonators we previously used in our studies of ammonia (Joiner *et al.*, 1989 and Joiner and Steffes, 1991). These measurements will initially be conducted in the frequency range from 32-40 GHz (7.5-9.38 mm wavelengths), which corresponds to the frequency of the new "Ka-Band" radio occultation measurement which will be conducted from the Cassini orbiter at Saturn. Subsequently, measurements in the 3 mm (100 GHz) range will be conducted, in support of new mapping efforts of the outer planets to be conducted with various arrays (BIMA and MMWA).

C. Applications to Observations and Radiative Transfer Modelling

Once the new formalism for the *microwave* (i.e. < 30 GHz) opacity of phosphine under conditions for the outer planets is completed (in the second year of the proposed three year project), the results will be applied to the interpretation of existing results from the Voyager radio occultation measurements at all four of the Jovian planets, as well as to the results from the Galileo entry probe radio opacity measurements, in order to determine the role of PH_3 in the microwave opacity of those atmospheres, and to possibly determine its deep atmospheric abundance. The formalism will also be applied to our radiative transfer model for Neptune (DeBoer and Steffes, 1996a), so as to derive accurate estimates of PH_3 abundance from Neptune microwave emission data. In the third year of the project, we will attempt to integrate our millimeter-wavelength laboratory results into an extended frequency (1-300 GHz, or 1 mm - 30 cm) formalism for phosphine opacity under outer planetary conditions. This will be timely in assisting pre-encounter planning of the Cassini radio science experiments at Saturn, and in possible complementary imaging of Saturn at millimeter-wavelengths from Earth-based telescope arrays.

IV. Proposed Procedure and Level of Effort

The proposed level of effort for the scientific research (not including Education/Public Outreach) in the 3 year period proposed (January 1, 1999 through December 31, 2001) involves one professor (P.G. Steffes, Professor of Electrical and Computer Engineering) at 25% time, and one graduate student (Graduate Research Assistant, James P. Hoffman) at 50% time, with supplies and other support as indicated in the attached cost breakdown. (See pp iii-vi, in the front section of this proposal.) (Note that 50% is the maximum support level for Ph.D. students, with the remaining 50% considered as registered academic thesis research.) In addition to the participation in the program by Professor Steffes and the paid graduate research assistant, contributions to the program from both

graduate and undergraduate students working on special projects for academic credit have been substantial. Likewise, in the spirit of the NASA Graduate Student Researchers Program, and in conjunction with Georgia Space Grant Consortium, we continue to seek out talented underrepresented minority students and involve them in our program.

V. Facilities

The specific laboratory measurements described in this proposal will be conducted at the Planetary Atmospheres Laboratory and the accompanying Remote Sensing Laboratory, which are located within the School of Electrical and Computer Engineering at Georgia Tech. A description of the equipment being used for these measurements is given in Section III.B of this proposal, and in the appended papers.

For support of the required data analysis and computing activities, a wide range of computing services for education, research, and administration is provided by the Georgia Tech Office of Information Technology. Numerous personal computers are also available to support this project.

VI. References

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VII. EDUCATION AND PUBLIC OUTREACH PROPOSAL

“Parent” Project: Laboratory Evaluation and Application of Microwave Absorption Properties under Simulated Conditions for Planetary Atmospheres.

Principal Investigator: Paul G. Steffes (Georgia Institute of Technology)

Grant Number: NAG5-4190

E/PO Project Title: Exploring the Solar System

A. ABSTRACT

The Georgia Tech Woodbury Research Facility contains the largest microwave antennas (two antennas, each 30-meter diameter) in the Southeastern United States. These antennas have been recently renovated for use in radio astronomy, spacecraft tracking, and SETI (not under NASA support). In a different E/PO proposal, D.R. DeBoer proposes to use this facility to introduce students to aspects of radio astronomy. In this complementary proposal, we will use the facility to focus on solar system exploration, starting with simple lessons on sizes of solar system bodies, and the extreme distances involved, going up to the level of actually receiving a signal from one of the active solar system exploring spacecraft, such as Pioneer 6, Pioneer 10, Voyagers 1 and 2, Ulysses, Mars Global Surveyor, or Cassini. This project will be conducted by the Principal Investigator, two Undergraduate Assistants, and a contractor who aids with site operation. It will focus on middle school students (grades 6-8), who have been shown to be the best target audience for introduction to space science. The resources of the Georgia Space Grant Consortium, based here at Georgia Tech will be used to actually conduct the teacher and student programs, thus allowing the principals supported by this E/PO to focus on the science education aspects of the projects.

B. SUMMARY

While American primary students continue to perform well on international science and mathematics tests, the performance of U.S. secondary students rates among the lowest in the industrial world. This E/PO project focuses on middle school students (grades 6-8) and their teachers, since this group appears to have the best potential to benefit from interactive science experiences. This project will invite middle school classes from throughout Georgia (with transportation arranged by the Georgia Tech Space Grant Consortium) to visit the Georgia Tech Woodbury Research Facility, a 40-acre site which includes a 10,000 square-foot building, and two 30-meter diameter parabolic dish antennas. When the students arrive, they are struck by the large size of the antennas, and they receive an brief initiation to the concept of the massive nature of the endeavour which is solar system exploration. After they arrive, the large antenna is pointed toward the horizon, so that its massive nature becomes apparent. They then walk along a path of disks, painted to represent the various planets, while being told that the 30-meter antenna

itself represents the relative size of the sun. Like the famous "Powers of Ten" film, this dramatically reinforces the massive size of the sun, and the relative smallness of the terrestrial planets as they walk by a 3-meter diameter Jupiter, and later pass a one-foot diameter earth. The students will then be led to a different part of the property, where the sun will now be scaled to a 0.3 meter size, and the relative distances to the nearly microscopic planets can be walked. (The walk to the orbits of Neptune and Pluto will be nearly 200 meters!) The largeness of the solar system and the miniscule size of the planets will serve as a very good starting point for discussing how challenging the discovery of planets in other solar systems might be. It also will serve to show the challenge of planetary missions within our own solar system.

After naming and briefly describing the various solar system exploration missions, the antenna will be pointed toward one of the spacecraft, and the students will actually see on a spectral display the weak signal from the spacecraft. They will then get a chance to ask questions and will learn of the basic limitations (such as the rotation and orientation of the earth) on earth tracking stations. At the end of the 3 hour experience (there will be a break for lunch as well), the students will leave with a set of discussion questions and an e-mail address to send questions.

C . STAFFING

While many university professors can be very good at explaining scientific concepts to general audiences, we will take special advantage of the large number of undergraduates in our program at Georgia Tech who are interested both in space exploration and in education. Experience has shown that the grades 6-8 students relate better to undergraduate students than to older adults, and the availability for this program of both female undergraduates and minority undergraduates will provide an important outreach to groups traditionally underrepresented in the physical sciences and engineering. The lesson plans and coordination will be handled by the Principal Investigator, but the presentations will be made by the undergraduate students. Coordination with the middle schools, scheduling, and transportation will be handled by the Georgia Space Grant Consortium.

D. TARGETS LEVELS

Given the limited size of this program, plus other uses for the facility, there will be 8 days available per calendar year for this activity. We will target student groups in the range of about 60-70 students. Thus, we expect to directly influence about 500 students per year. At a cost to the E/PO program of approximately \$20 per student, this represents an excellent investment.

E. BUDGET

For the period from January 1, 1999 through December 31, 2001:

	<u>CY99</u>	<u>CY00</u>	<u>CY01</u>
1. <u>Direct Labor*</u>	<u>\$5,657</u>	<u>\$5,657</u>	<u>\$5,657</u>
A. Principal Investigator (Paul G. Steffes)			
2% time, calendar year, (.02 person-years)	\$2,282		
B. Student Assistants (undergraduate)			
17.4% time, calendar year (.17 person-years)	\$2,789		
C. Fringe benefits (25.7% of direct salaries and wages, less students)	\$ 586		
2. <u>Other Direct Costs</u>			
a. Subcontract	<u>\$1,050</u>	<u>\$1,050</u>	<u>\$1,050</u>
(Site attendant, 4% time, calendar year (.04 person-years))			
3. <u>Facilities and Administrative Costs:</u>	<u>\$3,293</u>	<u>\$3,293</u>	<u>\$3,293</u>
Overhead (indirect expense), 49.1 % of direct cost base**			
4. <u>Total Estimated Costs:</u>	<u>\$10,000</u>	<u>\$10,000</u>	<u>\$10,000</u>

TOTAL FOR CY 99, 00, AND 01: \$30,000

*The salary and wage rates are based on FY99 salaries for the Georgia Institute of Technology. The Georgia Tech Fiscal Year is July 1 through June 30.

**Rates are for the period July 1, 1998 through June 30, 1999, and are subject to adjustment upon DCAA audit and ONR negotiations.

VIII. BIOGRAPHICAL SKETCH

PAUL G. STEFFES
PROFESSOR
SCHOOL OF ELECTRICAL AND COMPUTER ENGINEERING
GEORGIA INSTITUTE OF TECHNOLOGY
ATLANTA, GEORGIA 30332-0250

EDUCATION

S.B. Electrical Engineering 1977 Massachusetts Institute of Technology
S.M. Electrical Engineering 1977 Massachusetts Institute of Technology
Ph.D. Electrical Engineering 1982 Stanford University

EMPLOYMENT HISTORY

---Massachusetts Institute of Technology, Research Laboratory of Electronics, Radio Astronomy and Remote Sensing Group: Graduate Research Assistant, 1976-1977
---Watkins-Johnson Company, Sensor Development, San Jose, California Member of the Technical Staff, 1977-1982.
---Stanford University, Electronics Laboratory, Center for Radar Astronomy, Stanford, California, Graduate Research Assistant, 1979-1982.

Georgia Institute of Technology, School of Electrical and Computer Engineering,
Atlanta, Georgia

Assistant Professor	1982-1988
Associate Professor	1988-1994
Professor	1994-Present

RESEARCH EXPERIENCE SUMMARY (At Georgia Tech):

Principal Investigator of the National Science Foundation Grant, "Remote Sensing of Clouds Bearing Acid Rain." This research studied and designed a microwave/millimeter-wave system for remotely sensing the pH of acidic clouds (1982-1983). Principal Investigator of the NASA Planetary Atmospheres Program, "Laboratory Evaluation and Application of Microwave Absorption Properties Under Simulated Conditions for Planetary Atmospheres." This research includes study of the interaction between atmospheric constituents and electromagnetic waves, along with application of these studies to spacecraft and radio telescopic measurements of the microwave absorption in atmospheres of Venus and the outer planets (1984-1998). Principal Investigator of the GTE Spacenet Program, "Satellite Interference Locating System (SILS)." The program involved location of uplink signals on the surface of the earth without disrupting regular satellite operations (1986-1990). Principal Investigator of the Emory University/Georgia Tech Biomedical Technology Research Center project, "Research in Development of a Non-Invasive Blood Glucose Monitoring Technique." This research involved the use of active infrared systems to determine glucose levels in the human eye and bloodstream (1988-1989), with subsequent support (1990-1991) from Lifescan, Inc. Principal Investigator of the NASA Pioneer Venus Guest Investigator Program, "Pioneer Venus Radio Occultation (ORO) Data Reduction: Profiles of 13 cm Absorptivity." This research

inferred 13 cm wavelength absorptivity profiles using the Pioneer Venus Orbiter, and then used such profiles to characterize abundance profiles for gaseous H₂SO₄ in the Venus atmosphere (1988-1990). Principal Investigator/Team Member of NASA High Resolution Microwave Survey (HRMS). This research involved development and operation of the world's most sensitive receiving system used for a 1-10 GHz Sky Survey (1991-1994). Subsequent support has been provided by the SETI Institute (1994-1998). Developer of atmospheric radio occultation experiments conducted with the Magellan (Venus) Spacecraft (1991-1994). Director of the Ku Band Satellite Earth Station System. Responsible for development of a Ku-band uplink/downlink system for use in inter-university networks (1985-1995). Principal investigator in the NASA/ACTS Propagation Experiments Program (1994-1998). This research involves study of Ka-Band propagation effects.

Teaching Activities: Resource Professor for "Satellite Communications Systems" (graduate course). "Electromagnetics Applications" (undergraduate course covering Smith Charts, waveguides, and antennas), have also taught "Electromagnetics II (electrodynamics)", "Signals and Systems," and "Survey of Remote Sensing."

HONORS AND AWARDS

Member, Eta Kappa Nu; Member, Sigma Xi; Senior Member, IEEE (Member of 6 IEEE Societies).

Recipient of the Metro Atlanta Young Engineer of the Year Award, presented by the Society of Professional Engineers, 1985.

Recipient of the Sigma Xi Young Faculty Research Award, 1988.

Associate Editor, Journal for Geophysical Research (JGR-Atmospheres), 1984-1989.

Appointed Member of the NASA Management and Operations Working Group for the Planetary Atmospheres Program (1986-1990).

Elected to the Electromagnetics Academy, October 1990.

Recipient of the Sigma Xi Best Faculty Paper Award, 1991.

Recipient of the NASA Group Achievement Award, "For outstanding contribution to the design, development, and operation of the High Resolution Microwave Survey Project, and its successful inauguration," March 1993.

Recipient of the 1996 IEEE Judith A. Resnik Award, "For contributions to an understanding of the Venus atmosphere through innovative microwave measurements," January 1996.

OTHER PROFESSIONAL AFFILIATIONS

Member, American Association for the Advancement of Science.

Member, American Astronomical Society, Division for Planetary Sciences.

Member, American Geophysical Union; Member, American Institute of Physics.

Member, American Society for Engineering Education.

Elected Member, International Union of Radio Scientists (URSI), Commission J (Radio Astronomy).

OTHER PROFESSIONAL ACTIVITIES

Proposal Reviewer for the NASA Planetary Astronomy Program, the NASA Planetary Atmospheres Program, the NASA Planetary Instrument Definition and Development Program, the NASA planetary Data Analysis Programs, the NASA Exobiology Program, and the NSF Communications Research Program.

Reviewer/Referee for *Icarus* (International Journal of Solar System Studies), *Journal of Geophysical Research*, *Radioscience*, *IEEE Microwave and Guided Wave Letters Science*, and for several textbooks in the area of electromagnetics.

SELECTED RECENT PUBLICATIONS IN THE SPACE SCIENCES AREA:

- P. G. Steffes and D. R. DeBoer, "A SETI Search of Nearby Solar-Type Stars at the 203 GHz Positronium Hyperfine Resonance," *Icarus*, vol. 107, pp. 215-218, January, 1994.
- D. R. DeBoer, and P.G. Steffes, "Laboratory Measurements of the Microwave Properties of H₂S under Simulated Jovian Conditions with an Application to Neptune", *Icarus*, Vol. 109, pp. 352-366, June 1994.
- P. G. Steffes, J. M. Jenkins, R. S. Austin, S. W. Asmar, D. T. Lyons, E. H. Seale, and G. L. Tyler, "Radio Occultation Studies of the Venus Atmosphere with the Magellan Spacecraft. 1. Experiment Description and Performance," *Icarus*, vol. 110, pg. 71-78, July 1994.
- J. M. Jenkins, P. G. Steffes, J. Twicken, D. P. Hinson, and G. L. Tyler, "Radio Occultation Studies of the Venus Atmosphere with the Magellan Spacecraft 2. Results from the October 1991 Experiment" *Icarus*, vol. 110, pg. 79-94, July 1994.
- D. R. DeBoer and P. G. Steffes, "The Georgia Tech High Sensitivity Microwave Measurement System," *Astrophysics and Space Science*, Vol. 236, pp111-124, February 1996.
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B. Pending Support (other than this proposal):

NONE

The Microwave Absorption and Abundance of Sulfuric Acid Vapor in the Venus Atmosphere Based on New Laboratory Measurements

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New abundance profiles of gaseous sulfuric acid (H_2SO_4) have been computed based on the measured absorptivity profiles of the Venus atmosphere obtained during the 1991–1992 Magellan radio occultation experiments (Jenkins *et al.*, *Icarus* 110, 79, 1994; Hinson and Jenkins, *Bull. Am. Astron. Soc.* 27, 1079, 1995) and the 1974 Mariner 10 radio occultation experiment (Lipa and Tyler, *Icarus* 39, 192, 1979). The Magellan abundance profiles are different than those previously reported by Jenkins *et al.* due to the completion of new laboratory measurements of the microwave opacity of gaseous H_2SO_4 in a CO_2 environment. While these new experiments follow the same general procedure as Steffes (*Icarus* 64, 576, 1985; *Astrophys. J.* 310, 482, 1986), a more accurate determination of the H_2SO_4 to CO_2 number mixing ratio is obtained by using resonators with gold as opposed to silver plating to eliminate unaccounted-for chemical reactions. In addition, a significant reduction in the uncertainty of the absorptivities is achieved due to our ability to account for changes in the dielectric properties of the resonators when a lossy gaseous mixture is introduced into them. New power law multiplicative expressions for the microwave opacity of gaseous H_2SO_4 in a CO_2 environment are presented. The opacity expressions at S- and X-band are then applied to the Mariner 10 and Magellan absorptivity profiles, yielding sub-cloud gaseous H_2SO_4 abundances ranging from 10–14 ppm at the equator to 3–7 ppm at 67° latitude to 6–10 ppm at 88° latitude. The differences between profiles indicate both zonal and latitudinal variations in H_2SO_4 vapor abundance. © 1998 Academic Press

Key Words: Venus; atmospheres; abundances; experimental techniques.

1. INTRODUCTION

A significant number of data on the Venus atmosphere have been obtained in recent years as a result of the Magellan radio occultation experiments. In particular, new absorptivity profiles of the Venus atmosphere at 13 cm (2.298 GHz or S-band) and 3.6 cm (8.426 GHz or X-band) have been inferred from the October 1991 occultation experiments (Jenkins *et al.*, 1994). The $1-\sigma$ error bars of these absorptivity measurements are on the order of 10 to 15%, as compared to error bars on the order of 80% from similar measurements obtained from the Pioneer-Venus occultation experiments (Jenkins and Steffes, 1991).

The primary contributors to the microwave absorption in the Venus atmosphere are carbon dioxide (Ho *et al.*, 1966), sulfur dioxide (Janssen and Poynter, 1981) and sulfuric acid vapor (Steffes and Eshleman, 1981). The microwave opacity of carbon dioxide (CO_2) due to collisionally induced dipole moments is well known (Ho *et al.*, 1966). New laboratory measurements of the microwave absorption of sulfur dioxide (SO_2) under simulated Venus conditions have recently been performed to accuracies better than 10% (Suleiman *et al.*, 1996). Earlier laboratory measurements by Steffes (1985, 1986) indicate that the dominant microwave absorber at S- and X-band in the 10 km region below the Venus cloud layer (approximately 38–48 km) is sulfuric acid vapor (H_2SO_4). The Magellan radio occultation results exhibit the largest absorptivities in this region. The gaseous H_2SO_4 thermally dissociates into SO_3 and H_2O at altitudes between 35 and 40 km.

However, the laboratory measurements performed by Steffes (1985, 1986) of the microwave absorption of H_2SO_4 vapor under simulated Venus conditions have error bars

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on the order of 50%. Moreover, the ratio of the measured X-band to S-band absorptivities (≈ 10 to 1) does not match that measured from the October 1991 Magellan radio occultation experiments (≈ 4 to 1). Therefore, different abundance profiles of H_2SO_4 vapor are obtained when the multiplicative expressions for the S- and X-band absorptivities from Steffes (1985) are applied. The profiles of gaseous H_2SO_4 abundance in Jenkins *et al.* (1994) were derived using the S-band multiplicative expression from Steffes (1985). The abundances reached as high as 24 ppm and required significant supersaturation at the cloud base when compared to the H_2SO_4 saturation abundance. One explanation for this discrepancy is that the measurements of Steffes (1985) were performed using a silver-plated cylindrical cavity resonator which gave rise to unaccounted-for chemical reactions between the silver and the acid. The H_2SO_4 number mixing ratio was therefore overestimated and the expressions for microwave absorption normalized to mixing ratio were understated. Thus, larger abundances of gaseous H_2SO_4 were required to match the absorptivities measured in the Venus atmosphere.

Due to the large uncertainties and inconsistencies between the previous laboratory results and the Magellan radio occultation results, new laboratory measurements of the microwave opacity of H_2SO_4 vapor in a CO_2 environment have been completed at four wavelengths in the pressure range from 1 to 5 atm, which corresponds to the region in the Venus atmosphere where the gaseous H_2SO_4 resides. In this paper, we describe the experimental apparatus and procedure, highlight the changes and improvements made since the experiments of Steffes (1985, 1986), and present results for the gaseous H_2SO_4 pressure broadened microwave absorptivity. Best fit models of our data are then applied to the October 1991 Magellan radio occultation results to yield new abundance profiles of H_2SO_4 vapor in the Venus atmosphere. Finally, latitudinal variations in gaseous H_2SO_4 abundance are inferred by comparing the October 1991 profiles (at a latitude of 67°N) with profiles derived from the December 1992 polar Magellan results (Hinson and Jenkins, 1995) and the equatorial Mariner 10 radio occultation results (Lipa and Tyler, 1979).

2. EXPERIMENTAL APPARATUS

The experimental setup shown in Fig. 1 consists, first, of a Venus atmosphere simulator subsystem. The high temperatures on Venus are simulated using a temperature chamber capable of reaching temperatures as high as 600 K. The temperature chamber is regulated by a digital temperature controller and monitored by an iron-constantan (J-type) thermocouple attached to a digital voltmeter (dvm). The reference junction of the thermocouple is embedded in a cold bath to insure that the voltage reading on the dvm is dependent only on the temperature where the iron and constantan tips are in contact.

The pressures characteristic of the Venus atmosphere are contained inside a cylindrical stainless steel (type T304) pressure vessel, capable of withstanding pressures of up to 6 atmospheres, which is placed inside the temperature chamber. The pressure vessel is 46 cm in height and 23 cm in radius. A Viton O-ring is compressed between the lid and top of the pressure vessel with stainless steel hex nuts. The pressure vessel lid also contains a probe which extends 5 cm down into the pressure vessel. The tip of the J-type thermocouple is suspended down to the base of the probe so that it correctly monitors the temperature inside the pressure vessel.

A vacuum in the pressure vessel is attained using a vacuum pump capable of achieving vacuums of 1–2 Torr. The vacuum state is monitored by a digital thermocouple vacuum gauge which can measure pressures in the range from 1 to 800 Torr. Likewise, positive pressure from a cylinder of high purity carbon dioxide is monitored by an analog pressure gauge which can measure pressures in the range from 1 to 7.5 atm. Connected to the pressure vessel inside the temperature chamber is a Pyrex glass flask which contains the liquid sulfuric acid (98.65% concentration by weight). All elements of the subsystem are connected by a network of 3/8" stainless steel tubing with Swagelok fittings. Valves are also placed throughout the network so that parts of the subsystem may be isolated to detect leaks and insure safety.

The second subsystem, the microwave measurement subsystem, consists of two gold-plated circular cylindrical cavity resonators. These resonators were also used in the experiments of Steffes (1985, 1986), but with silver as opposed to gold plating. The larger of the two resonators has a radius of 13.10 cm and a height of 25.32 cm and is placed at the base of the pressure vessel. The dimensions of this resonator are optimal for measurements in the 2 to 9 GHz (S- and X-band) frequency range. Energy is coupled into and out of the cavity by input and output ports containing adjustable BNC jack to jack adapters with closed loop signal probes attached. The smaller of the two resonators has a radius of 2.475 cm and a height of 4.850 cm and rests on a shelf which hangs from the lid of the pressure vessel. The dimensions of this resonator are optimal for measurements in the 10 to 26 GHz (KU- and K-band) frequency range. Energy is similarly coupled into and out of the cavity by input and output ports containing adjustable SMA jack to jack adapters with open loop signal probes attached. Each resonator also contains two thin slits which serve the dual purpose of suppressing degenerate low quality factor TM resonant modes and allowing gas to flow freely into the resonators.

The input ports of each cavity resonator are connected to one of two HP 8690B microwave sweep oscillators. Each sweeper consists of a mainframe plus a replaceable backward wave oscillator (BWO) module. S-band (2–4 GHz) and X-band (8–12 GHz) BWO modules are used for input

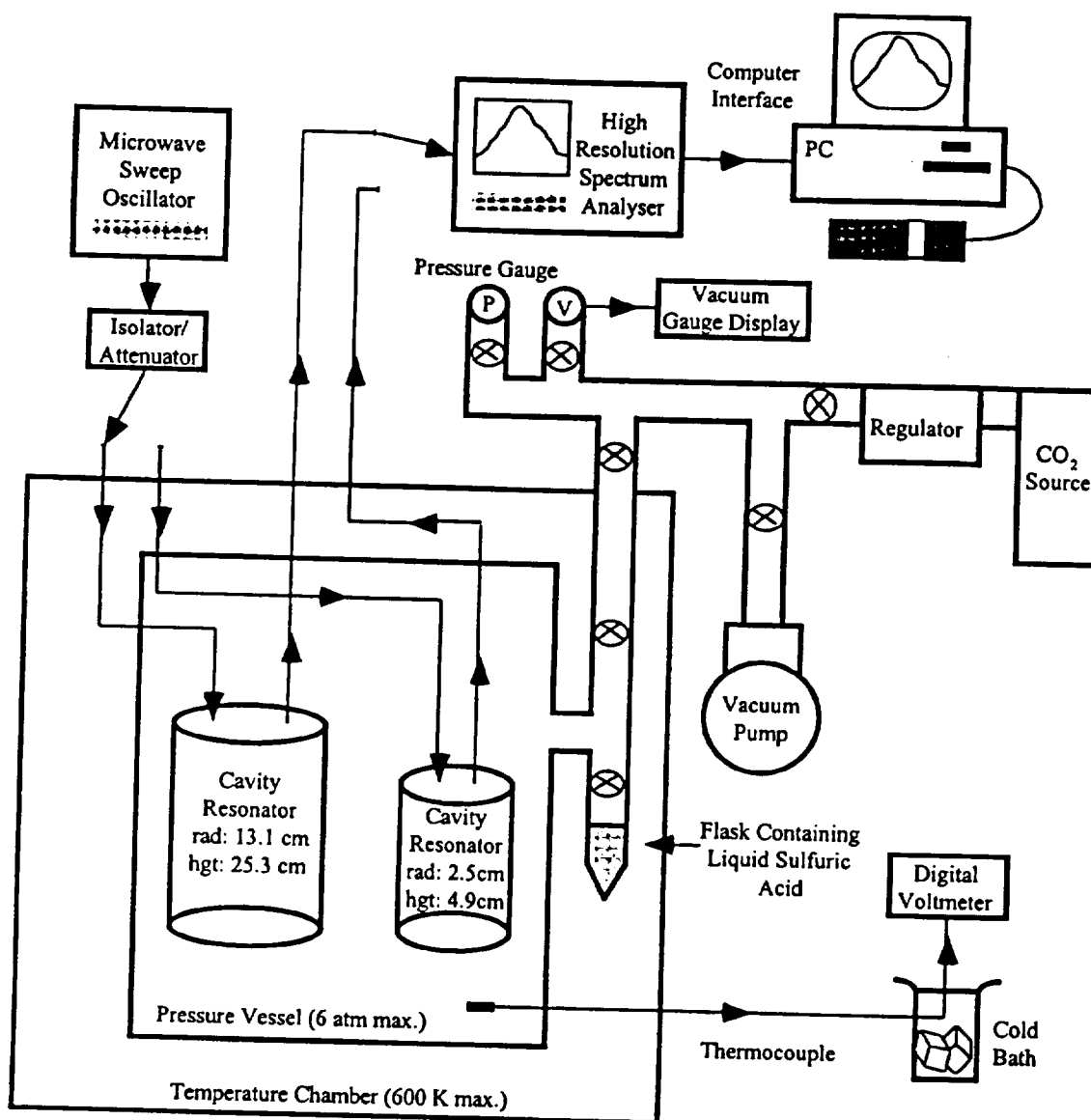


FIG. 1. Block diagram of the experimental setup, as configured for measurements of the microwave absorption of gaseous sulfuric acid under Venus atmospheric conditions.

to the large resonator while KU-band (12–18 GHz) and K-band (18–26 GHz) BWO modules are used for input to the small resonator. The signal from the output port of each resonator is displayed on a high resolution HP 8562B spectrum analyzer. The output terminals of the S- and X-band BWO modules are connected to S- and X-band ferrite isolators while the output terminals of the KU- and K-band BWO modules are connected to a 3 dB attenuator to reduce any reflected signal from the resonators back toward the sweep oscillators.

Flexible coaxial cables with BNC connectors are used to and from the ports of the large resonator both inside and outside the pressure vessel. Two N-type hermetically sealed bulkhead feedthroughs with N-type to BNC jack

adapters are attached to the pressure vessel lid. Likewise, custom-made semi-rigid coaxial cables with silicone dioxide dielectric and SMA connectors are used to and from the ports of the small resonator inside the pressure vessel. Two SMA hermetically sealed bulkhead feedthroughs are attached to the pressure vessel lid, which are in turn connected to splined semi-rigid coaxial cables with SMA connectors outside the pressure vessel.

The final subsystem is a data acquisition subsystem consisting of a personal computer and a software package which interfaces with the HP 8562B spectrum analyzer via a general purpose interface bus (GPIB). Six hundred data points from each sweep of a resonance on the spectrum analyzer are read into the computer and smoothed. The

amplitude, center frequency, and half-power bandwidth of the resonance are then recorded. Statistical data is collected as a series of sweeps are performed on a resonance and averaged. A complete discussion of the data acquisition subsystem can be found in DeBoer and Steffes (1996). The addition of this subsystem has greatly improved the sensitivity and reliability of this measurement system as compared to the experiments of Steffes (1985, 1986) where the operator was required to eye-fit the center frequency and half-power bandwidth directly from the spectrum analyzer.

3. EXPERIMENTAL APPROACH

3.1. Measurement of the Absorptivity

A useful technique for computing the microwave absorptivity of a gaseous mixture at a particular frequency is to measure the change in the quality factor and amplitude of a cylindrical cavity resonance at that frequency when the gas mixture is introduced into the resonator (Townes and Schawlow, 1955). A convenient relation between the quality factor change and the absorptivity is derived by DeBoer and Steffes (1994),

$$\alpha = 4.343 \frac{2\pi}{\lambda} \left(\frac{1 - \sqrt{t_{\text{mix}}}}{Q_{\text{mix}}} - \frac{1 - \sqrt{t_{\text{ref}}}}{Q_{\text{ref}}} \right) \frac{\text{dB}}{\text{km}}, \quad (1)$$

where λ is the wavelength in kilometers at the center of the resonance with the gas mixture present and Q and t are the measured quality factor and transmissivity of the resonator with the gas mixture (mix) or reference lossless gas (ref) present. The quality factor Q is simply the ratio of the cavity resonant center frequency to its half-power bandwidth and the transmissivity t is measured directly from the amplitude of the resonance.

Note that a lossless gas is used as a reference for measuring the change in the quality factor of a particular resonance. The reference quality factors in the experiments of Steffes (1985, 1986) were measured under a vacuum. When a gas mixture (absorbing or nonabsorbing) is introduced into a cavity resonator, though, the real (lossless) part of its dielectric constant can alter the quality factor of the cavity itself (Q_c). This effect is known as dielectric loading. To correctly reference a resonance measurement under mixture pressure, an appropriate amount of a lossless gas (in our case pure CO_2) is added so that the shift in the center frequency of the resonance from vacuum is the same as the shift when the gas mixture was introduced into resonator. The refractive indices of the mixture and the reference lossless gas are then matched which compensates, in principle, for the effects of dielectric loading. Therefore, the center frequencies of the resonance under the gas mixture and the reference gas are equal, and Eq. (1) can be rewritten as:

$$\alpha = 4.343 \frac{2\pi}{v_{\text{mix}}} ((1 - \sqrt{t_{\text{mix}}})\Delta f_{\text{mix}} - (1 - \sqrt{t_{\text{ref}}})\Delta f_{\text{ref}}) \frac{\text{dB}}{\text{km}}, \quad (2)$$

where Δf_{mix} and Δf_{ref} are the half-power bandwidths in the presence of the gas mixture and reference gas. v_{mix} , the speed of light in the gas mixture, is defined as:

$$v_{\text{mix}} = c f_{\text{mix}} / f_{\text{vac}}, \quad (3)$$

where f_{mix} and f_{vac} are the center frequencies of the resonance under the gas mixture and under vacuum, and c is the speed of light in a vacuum.

3.2. Measurement of the Mixing Ratio

In order to establish a useful expression for opacity, the absorptivity measurements must be normalized to the H_2SO_4 number mixing ratio in the $\text{CO}_2/\text{H}_2\text{SO}_4$ mixture. The mixing ratio was the primary source of uncertainty in the experiments of Steffes (1985, 1986) due to reactions between the acid vapor and the silver plating on the resonators. Great care was taken for these new laboratory measurements to eliminate any potential source of reaction. As mentioned previously, the silver plating was removed from the resonators and replaced with gold plating. The pressure vessel, the 3/8" tubing, the shelf which the small resonator sits on, and the supporting wires are stainless steel. Likewise, the custom-made semi-rigid coaxial cables inside the pressure vessel are externally coated with a layer of stainless steel.

The number mixing ratio is computed by measuring the volume of liquid H_2SO_4 which is vaporized into the pressure vessel under vacuum. The operating temperature for these new measurements is 553 K (280°C). This temperature is high enough so that a sufficient amount (greater than 3 ml) of H_2SO_4 solution will vaporize, but low enough so that the Viton O-ring and the hermetic seals in the bulkhead feedthroughs will not leak. The original concentration of the H_2SO_4 solution is 98.65% by weight. Once vapor pressure equilibrium is attained, the H_2SO_4 concentration in the liquid phase reaches the azeotropic concentration under vacuum of 98.9% (Kunzler, 1953). From the volume of the H_2SO_4 solution which vaporizes, the known density of pure H_2SO_4 liquid (1.8305 g/ml), and the molecular weight of H_2SO_4 (98.08 g/mole), the number of moles of pure H_2SO_4 liquid which vaporizes, n_{vap} , can be computed. Note that n_{vap} does not include the free water which vaporizes from the solution. Once in vapor phase, though, some of the H_2SO_4 molecules will thermally dissociate into H_2O and SO_3 . In particular,

$$n_{\text{H}_2\text{SO}_4} = n_{\text{vap}} (1 - D), \quad (4)$$

where $n_{\text{H}_2\text{SO}_4}$ is the number of moles of H_2SO_4 vapor and Δ , the dissociation constant, is defined as:

$$D = P_{\text{SO}_3} / (P_{\text{H}_2\text{SO}_4} + P_{\text{SO}_3}), \quad (5)$$

where $P_{\text{H}_2\text{SO}_4}$ and P_{SO_3} are the partial pressures of H_2SO_4 and SO_3 .

The dissociation constant is dependent on the temperature and the concentration of the original solution. For a temperature of 553 K and a 98.65% by weight solution, the dissociation constant is equal to 0.461 (Vermeulen, 1984); thus less than half of the H_2SO_4 molecules dissociate. It should be noted that the microwave absorption of the dissociates, H_2O and SO_3 , is negligible in these small amounts when compared to the microwave absorption of H_2SO_4 vapor. (See, e.g., Ho *et al.*, 1966, for the microwave opacity of H_2O and Steffes and Eshleman, 1981 for an upper limit on the microwave opacity of SO_3 .) The vapor pressure of the remaining H_2SO_4 can then be computed from the known volume of free space in the pressure vessel (31 liters) and Eq. (4) using the ideal gas law. The number mixing ratio is simply the ratio of the H_2SO_4 vapor pressure to the total pressure inside the pressure vessel.

3.3. Measurement of the Refractivity

In addition, the refractivity of gaseous H_2SO_4 can be determined, following vaporization of the liquid into the evacuated pressure vessel, by measuring the change in the center frequency of a particular resonance relative to vacuum. The refractive index is simply the ratio of the resonant frequency at vacuum, f_{vac} , to the resonant frequency with the H_2SO_4 vapor present, f_{gas} . The refractivity N is defined as:

$$N = 10^6(n - 1) = 10^6(f_{\text{vac}} - f_{\text{gas}})/f_{\text{gas}}. \quad (6)$$

The measured refractivity N_{meas} is the sum of the refractivity of gaseous H_2SO_4 , its dissociates, and the free H_2O which vaporized from the solution along with the H_2SO_4 molecules:

$$N_{\text{meas}} = N'_{\text{H}_2\text{SO}_4} \rho_{\text{H}_2\text{SO}_4} + N'_{\text{SO}_3} \rho_{\text{SO}_3} + N'_{\text{H}_2\text{O}} \rho_{\text{H}_2\text{O}} \quad (7)$$

$$= N_A (N'_{\text{H}_2\text{SO}_4} n_{\text{H}_2\text{SO}_4} + N'_{\text{SO}_3} n_{\text{SO}_3} + N'_{\text{H}_2\text{O}} n_{\text{H}_2\text{O}}) / V_0,$$

where N' is number density normalized refractivity, ρ is the number density, n is the number of moles, N_A is Avogadro's number, and V_0 is the volume of free space in the pressure vessel (31 liters). The number of moles of H_2SO_4 is defined in Eq. (4). The number of moles of SO_3 and H_2O is

TABLE I
The Resonant Frequencies and Their Corresponding TE_{nm} Modes Used to Measure the Microwave Absorption of H_2SO_4 Vapor

Frequency band	Cavity resonator	Experiment number	Frequency (GHz)	TE_{nm} mode
S	large	1, 2	2.26	TE_{013}
X	large	1	8.39	TE_{0510}
		2	8.78	TE_{075}
KU	small	1, 2	11.89	TE_{013}
K	small	1, 2	21.61	TE_{033}

$$n_{\text{SO}_3} = (n_{\text{vap}} - n_{\text{H}_2\text{SO}_4}) = n_{\text{vap}} D, \quad (8)$$

$$n_{\text{H}_2\text{O}} = n_{\text{SO}_3} + n_{\text{free H}_2\text{O}}. \quad (9)$$

The normalized refractivities N' of SO_3 and H_2O (at 553 K) are

$$N'_{\text{SO}_3} = 1.156 \times 10^{-17} \text{ N-units cm}^3 \text{ molecule}^{-1}$$

(Steffes, 1982) and

$$N'_{\text{H}_2\text{O}} = 1.016 \times 10^{-16} \text{ N-units cm}^3 \text{ molecule}^{-1}$$

(Essen and Froome, 1951).

The normalized refractivity $N'_{\text{H}_2\text{SO}_4}$ is then computed from Eq. (7).

3.4. Selection of Resonances

Laboratory measurements of the microwave absorption of sulfuric acid vapor in a carbon dioxide environment were performed on two dates, namely June 5, 1996 (Experiment 1), and September 25, 1996 (Experiment 2). Prior to each experiment, a selection of resonances to be measured was made based initially on the proximity of the resonances to frequencies which had important applications such as the Magellan downlink frequencies. A second important criterion was the quality factor of the resonances. The narrower the half-power bandwidth of a resonance, the more sensitive we are to measuring changes in its quality factor. Other important criteria for selecting resonances are the signal-to-noise ratio and the symmetry of the resonances. As a final test, we add high pressures of a lossless gas (pure CO_2) to the pressure vessel, at room temperature, to determine whether or not the resonances are especially sensitive to dielectric loading. The frequencies of the resonances chosen for the experiments and their corresponding TE_{nm} modes are listed in Table I.

4. EXPERIMENTAL PROCEDURE

At the start of each experiment, the Pyrex glass flask is first filled with a precisely known volume (10 ml) of liquid sulfuric acid (98.65% by weight) at room temperature using a 1 ml syringe, and is attached to the pressure vessel via stainless steel tubing as shown in Fig. 1. The temperature chamber is turned on and the entire system is heated to a temperature of 553 K. The system takes approximately 6 h to reach thermal equilibrium. During this time period, the glass flask and the pressure vessel are isolated from each other and from the outside environment. As the resonators are heated, the resonances shift down in frequency due to thermal expansion. Once thermal equilibrium is reached, the resonant frequencies stabilize. Following the heating cycle, the H_2SO_4 solution is checked again to ensure that the acid has remained clear and that there is no evidence of contamination.

A vacuum is then drawn in the pressure vessel and the center frequencies, bandwidths, and amplitudes of the four resonances are measured. Next, the valve isolating the flask and the pressure vessel is opened, allowing the liquid sulfuric acid to vaporize into the pressure vessel. After about five minutes, vapor pressure equilibrium is reached. Shifts in the resonant frequencies are observed. The new center frequency of the S-band resonance is measured to determine the refractivity of the gaseous H_2SO_4 .

Carbon dioxide is then admitted into the pressure vessel at a slow rate (1 psig per minute) to minimize the risk of condensing the H_2SO_4 vapor. Once the designated pressure is reached and the resonances have stabilized (indicating pressure equilibrium within the resonators), the quality factor and amplitude of the four resonances under the H_2SO_4 - CO_2 mixture are measured. Subsequent pressures are obtained by either adding CO_2 or partially venting the mixture. During Experiment 1, the approach was to first produce a mixture at 5 atm and then vent to lower pressures. Note that 1 atm is equal to 1.013 bars. This method has the advantage that the mixing ratio (1.183%) remains the same at all pressures. During Experiment 2, the approach was to first produce a mixture at 1 atm and then add CO_2 to produce mixtures at higher pressures. While the mixing ratio at each pressure is different, this method has the advantage of increased sensitivity at the lower pressures due to a higher mixing ratio. The original mixing ratio, though, at 1 atm in Experiment 2 was 4.547%. Such a large mixing ratio could exhibit a significant amount of opacity due to self-broadening, as opposed to external broadening, which would be more difficult to model. Therefore, an additional 1 atm mixture was produced in Experiment 2 by venting from a higher pressure.

Once all measurements of the H_2SO_4 - CO_2 mixture are completed, a vacuum is drawn once again in the pressure vessel. The center frequency, bandwidth, and amplitude

of the resonances are measured again at this stage to insure consistency at vacuum. Pure CO_2 is then introduced into the pressure vessel to reference each gas mixture measurement. The proper amount of CO_2 has been added for matching a particular resonance measurement when the shift in the center frequency of the resonance relative to a vacuum is the same as the shift when the gas mixture was introduced into the resonator. Since the gas mixture is more refractive than pure CO_2 , a higher pressure of CO_2 is required for matching each gas mixture pressure. The quality factor and amplitude of the resonances are measured one last time. The system is then allowed to cool back down to room temperature. The remaining volume of the liquid H_2SO_4 in the flask (now at the azeotropic concentration of 98.9% by weight) is precisely measured. Volumes of 4.12 and 3.18 ml of sulfuric acid solution were vaporized in Experiments 1 and 2, respectively.

It may, at first, seem surprising that different volumes of the acid solution vaporized in each experiment since the operating temperature was the same, 553 K. Tests, though, have been performed (Fahd, 1992) which show that the vaporization of a small reservoir of H_2SO_4 solution (≤ 10 ml) into a vacuum from a narrow flask is not an accurate method for determining the saturation vapor pressure of H_2SO_4 . This was supported by the fact that a slight residue of sulfur (on the order of 0.02 ml) was always present at the bottom tip of the flask following the initial heating cycle before each experiment commenced. This residue, which is a natural byproduct of the heating process, may affect the amount of liquid solution which goes to vapor phase near the base of the flask. Note again that during the heating cycle, the glass flask was completely isolated from the pressure vessel and resonators. This small residue, though, should have a negligible effect on the overall concentration of the initial 10 ml H_2SO_4 solution. Our primary concern was to precisely measure the volume of vaporized solution so that the number mixing ratios could be accurately computed. At the conclusion of each experiment, both the resonators and the pressure vessel are carefully examined to ensure that there is no evidence of unaccounted-for chemical reactions between the acid vapor and the gold plating or stainless steel in the system.

5. UNCERTAINTY ANALYSIS

The quantities which are measured to compute the normalized absorptivity and refractivity of sulfuric acid vapor are subject to two classes of uncertainty, namely instrumental uncertainty due to the limitations of our measuring devices and statistical uncertainty due to random variations over a series of measurements of a particular quantity. First, the instrumental uncertainty of the system operating temperature (553 K) is based on the accuracy of the digital voltmeter, equal to $\pm[0.5\% \text{ of dvm reading} + 0.2 \text{ mV}]$,

TABLE II
Spectrum Analyzer Instrumental
Uncertainties at Resonant Frequencies

Frequency (GHz)	σ_f (kHz)	$\sigma_{\Delta f}$ (Hz)	σ_{amp} (dB)
2.26	48	602	1.5
8.39	139	805	1.5
11.89	385	4425	1.5
21.61	582	4839	1.5

yielding an error bar of ± 0.27 mV (for a 14.2 mV reading) or ± 5.4 K. The temperature measurement accuracy is confirmed from the reading on the digital temperature controller, which also monitors the temperature inside the pressure vessel via a second thermocouple.

An additional instrumental uncertainty arises from the limitations of the pressure gauge which consists of gradations 2 psig apart, corresponding to a pressure error bar of ± 0.136 atm. Likewise, the error bars of the number mixing ratio are based on the uncertainty in the volume of liquid H_2SO_4 which went to vapor phase. A 1 ml syringe with 0.01 ml gradations was used to add the original volume (10 ml) to the flask and to measure the remaining liquid at the conclusion of each experiment. Since 10 iterations are required to add acid to the flask and less than 10 iterations are required to measure the residual liquid, a conservative uncertainty of ± 0.2 ml is assigned for the volume of vaporized liquid H_2SO_4 . This sets the error bars on the mixing ratio, as well as on the number of moles of gaseous H_2SO_4 and its dissociates, which are used in the calculation of the H_2SO_4 vapor refractivity (Eq. (7)).

The calculation of the absorptivity error bars involves both instrumental and statistical uncertainty. The quantities required to compute the absorptivity at a particular pressure and frequency are, from Eqs. (2) and (3), the half-power bandwidth, the amplitude, and the center frequency of the resonance for the gas mixture and the reference gas. The instrumental errors in frequency, bandwidth, and amplitude are based on the limited accuracy of the spectrum analyzer used to measure these quantities. The standard deviations of these errors are shown in Table II as a function of frequency (Hewlett-Packard Corporation, 1986). The instrumental error of the center frequency is less than 0.01%, and is thus insignificant in calculating the speed of light in the gas mixture (Eq. 3). This error, though, is significant in the calculation of the gaseous H_2SO_4 refractivity (Eq. 6) error bar, where a very small change in the position of the S-band resonance center frequency is being measured. Similarly, the instrumental error of the amplitude yields a worst case uncertainty in the transmissivity t of less than 0.8%, and is therefore not included in the calculation of the absorptivity error bar.

At each pressure and frequency (for the gas mixture and the reference gas), 10 measurements of the resonant center frequency, bandwidth, and amplitude are recorded. Statistical variations due to random electrical noise occur. The variances of the center frequency and the amplitude, though, are less than 0.01% and 0.1%, respectively. The bandwidth measurements, on the other hand, exhibit significant statistical variations. The sample variance is computed for each set of bandwidth measurements and then multiplied by the confidence interval and normalized to the number of measurements to yield the true variance. A confidence interval of 1.88 is used, which corresponds to a 90% confidence level for 10 measurements of a data point (Papoulis, 1991). The total uncertainty of the absorptivity is then computed as:

$$\sigma_a = ((\sigma_{\text{mix}})^2 + (\sigma_{\text{ref}})^2)^{1/2}, \quad (10)$$

where the variances of the gas mixture and reference gas are:

$$(\sigma_{\text{mix}})^2 = \gamma_{\text{mix}}((\sigma_{\Delta f \text{ mix inst}})^2 + (\alpha_{\Delta f \text{ mix stat}})^2), \quad (11)$$

$$(\sigma_{\text{ref}})^2 = \gamma_{\text{ref}}((\sigma_{\Delta f \text{ ref inst}})^2 + (\alpha_{\Delta f \text{ ref stat}})^2 + (\sigma_{\Delta f \text{ match}})^2), \quad (12)$$

and:

$$\gamma_{\text{mix,ref}} = 4.343 \frac{2\pi}{v_{\text{mix,ref}}} (1 - \sqrt{t_{\text{mix,ref}}}). \quad (13)$$

The instrumental and statistical uncertainty in the bandwidth Δf have been discussed in the previous paragraphs. The final term in Eq. (12) characterizes the bandwidth uncertainty when the H_2SO_4 gas mixture is matched with the CO_2 reference gas. First, the high pressure mixtures (4 and 5 atm) could not be precisely matched due to the inability of our pressure seal at high temperatures to withstand the pressure levels required to match the high refractivity of the gas mixtures. Conversely, the lower pressure mixtures were always precisely matched, but in some cases, we noticed variations in the bandwidth of the reference gas with pressure, indicating the presence of dielectric loading. Conditions in the resonators later during the experiment, when the reference measurements were recorded, could be different than earlier when the gas mixture measurements were taken, despite being perfectly matched in terms of refractive index. We therefore add an extra term in Eq. (12) which characterizes the variation in the bandwidth of a particular resonance under the reference gas. To account for imperfect matching of the high pressure mixture data, variations in the reference bandwidth over the entire range of pressures are included. Likewise, for the lower pressure data, variations (if any) within a 1 atm range of the pressure where the reference gas is dielectrically matched to the gas mixture are included.

TABLE III
Laboratory Measurements of the Microwave Absorption of Sulfuric Acid Vapor at 553 K

Frequency (GHz)	Pressure (atm)	Experiment number	Number mixing ratio	Measured absorption (dB km ⁻¹)
2.26	5.0	1	0.0118 ± 0.0006	10.7343 ± 0.4898
2.26	3.5	1	0.0118 ± 0.0006	6.3812 ± 0.4935
2.26	3.0	2	0.0152 ± 0.0010	7.3911 ± 0.4140
2.26	2.0	2	0.0227 ± 0.0015	6.2259 ± 0.3886
2.26	1.0	2	0.0114 ± 0.0007	0.7330 ± 0.6831
8.39	5.0	1	0.0118 ± 0.0006	42.8675 ± 0.6860
8.39	4.0	1	0.0118 ± 0.0006	28.6222 ± 0.3353
8.78	3.0	2	0.0152 ± 0.0010	35.8383 ± 0.5290
8.78	2.0	2	0.0227 ± 0.0015	33.3905 ± 0.4864
8.39	1.0	1	0.0118 ± 0.0006	5.1979 ± 0.2633
8.78	1.0	2	0.0114 ± 0.0007	4.9484 ± 0.1421
11.89	5.0	1	0.0118 ± 0.0006	74.1878 ± 5.1503
11.89	3.4	2	0.0114 ± 0.0007	33.9563 ± 6.1511
11.89	2.0	2	0.0227 ± 0.0015	52.1787 ± 6.2500
11.89	1.0	1	0.0118 ± 0.0006	10.7582 ± 4.6205
11.89	1.0	2	0.0114 ± 0.0007	7.4194 ± 1.5613
21.61	5.0	1	0.0118 ± 0.0006	123.3030 ± 14.1908
21.61	3.0	2	0.0152 ± 0.0010	106.4710 ± 6.8188
21.61	1.0	1	0.0118 ± 0.0006	20.9695 ± 14.0351
21.61	1.0	2	0.0114 ± 0.0007	20.2955 ± 5.1313

6. EXPERIMENTAL RESULTS

The measured absorptivities α , number mixing ratios q , and 1- σ error bars from Experiments 1 and 2 are presented in Table III. The normalized absorptivities (α/q) are listed in Table IV and plotted in Figs. 2–5 as a function of pres-

sure for each frequency. Note that a frequency correction was performed on the X-band data from Experiment 2 in Table IV and Fig. 3 so that the results could be displayed with the data from Experiment 1. This correction is described in detail in Section 7. Figure 6 displays plots of the normalized absorptivity as a function of pressure for all

TABLE IV
Normalized Absorptivities of Sulfuric Acid Vapor at 553 K

Frequency (GHz)	Pressure (atm)	Measure α/q (dB km ⁻¹)	Model α/q (dB km ⁻¹)	Error (σ)
2.26	5.0	907.38 ± 61.29	918.56	-0.18
2.26	3.5	539.41 ± 48.54	566.32	-0.55
2.26	3.0	487.54 ± 41.72	459.50	0.67
2.26	2.0	273.79 ± 24.67	265.16	0.35
2.26	1.0	64.46 ± 45.53	103.59	-0.86
8.39	5.0	3623.63 ± 168.76	3605.96	0.10
8.39	4.0	2419.46 ± 105.33	2696.77	-2.63
8.39	3.0	2226.39 ± 133.18	1854.27	2.79
8.39	2.0	1382.88 ± 82.54	1093.71	3.50
8.39	1.0	439.39 ± 31.24	443.57	-0.13
8.39	1.0	409.88 ± 28.82	443.57	-1.17
11.89	5.0	6271.15 ± 529.21	6148.53	0.23
11.89	3.4	2986.48 ± 520.31	3695.58	-1.36
11.89	2.0	2294.58 ± 300.03	1834.38	1.53
11.89	1.0	909.40 ± 308.53	734.74	0.57
11.89	1.0	652.54 ± 127.22	734.74	-0.65
21.61	5.0	10422.87 ± 1216.49	11398.66	-0.80
21.61	3.0	7023.13 ± 640.88	6445.71	0.90
21.61	1.0	1772.57 ± 902.30	1891.49	-0.13
21.61	1.0	1785.00 ± 401.64	1891.49	-0.27

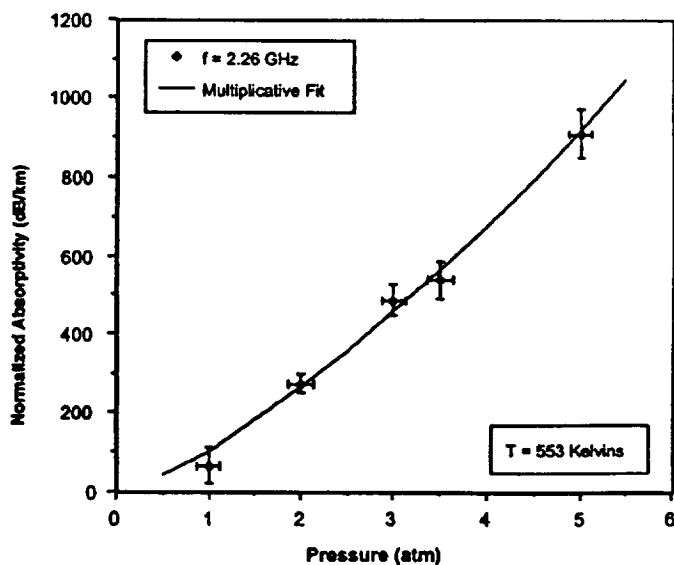


FIG. 2. Laboratory measurements of the absorption normalized to mixing ratio of sulfuric acid vapor as a function of pressure (1 atm = 1.013 bar) in a CO₂ environment at S-band.

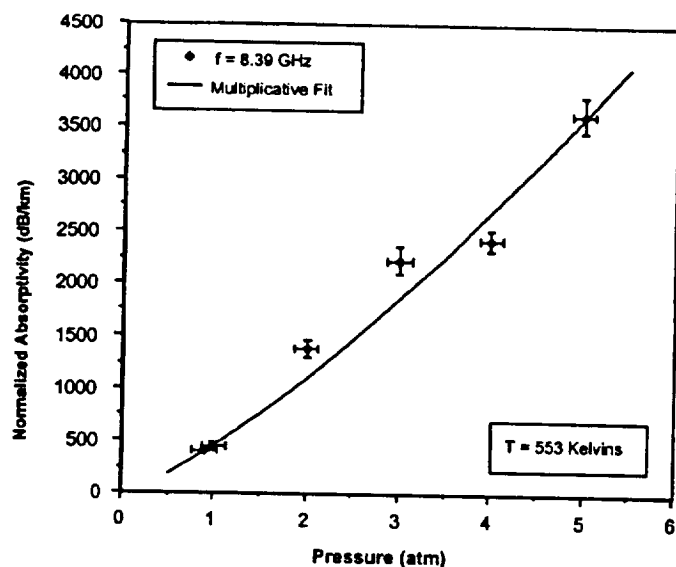


FIG. 3. Laboratory measurements of the absorption normalized to mixing ratio of sulfuric acid vapor as a function of pressure (1 atm = 1.013 bar) in a CO_2 environment at X-band.

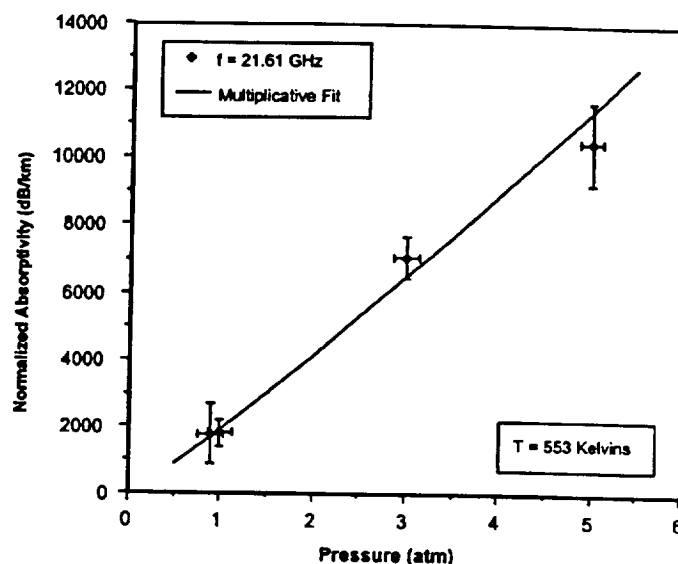


FIG. 5. Laboratory measurements of the absorption normalized to mixing ratio of sulfuric acid vapor as a function of pressure (1 atm = 1.013 bar) in a CO_2 environment at K-band.

frequencies. As expected, the new microwave opacities of pressure broadened sulfuric acid vapor are much higher than those reported by Steffes (1985, 1986). The primary reason for this, as described in the Introduction, was the use of gold as opposed to silver plated cavity resonators, which eliminated unaccounted-for chemical reactions between the acid vapor and the plating. In comparison to

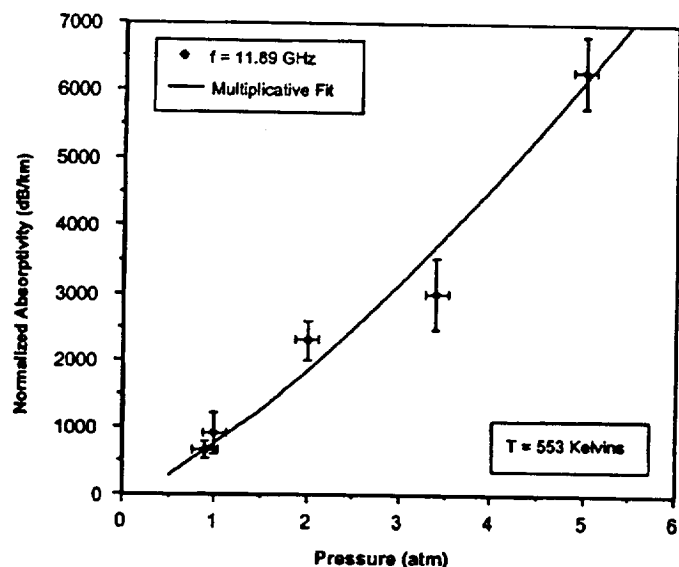


FIG. 4. Laboratory measurements of the absorption normalized to mixing ratio of sulfuric acid vapor as a function of pressure (1 atm = 1.013 bar) in a CO_2 environment at KU-band.

the measurements of Steffes (1985), the new normalized absorptivities are a factor of 5 to 7 times higher at S-band and a factor of 2 to 3 times higher at X-band. The ratio of X-band to S-band absorption has been reduced from roughly 10 to 1 to about 4 to 1. This differential change is primarily due to the presence of unaccounted-for dielectric loading in the Steffes (1985) measurements, which was much more pronounced at X-band than at S-band. This new ratio, as will be seen in Section 8, is much more consistent with the absorptivity profiles measured from the Magellan spacecraft. Finally, the measured number density normalized refractivities of sulfuric acid vapor from Exp. 1 and Exp. 2 are listed in Table V. The consistency between the normalized refractivities from each experiment support the measurements of and accuracies in the volume of vaporized sulfuric acid solution.

7. H_2SO_4 MICROWAVE OPACITY FORMALISM

To produce a microwave opacity formalism for pressure broadened H_2SO_4 vapor, we first attempted to model the laboratory data using the Poynter-Pickett-Cohen spectral line catalog for H_2SO_4 (Poynter and Pickett, 1985) with a Van Vleck-Weisskopf spectral lineshape. The intensities of the H_2SO_4 line centers were measured by Kuczkowski *et al.* (1981) in the 60 to 112 GHz frequency range. The line center intensities of the catalog extrapolated to centimeter wavelengths, though, were much too low to characterize the measured absorptivities. Therefore, the following power law multiplicative expressions are used to best fit the data,

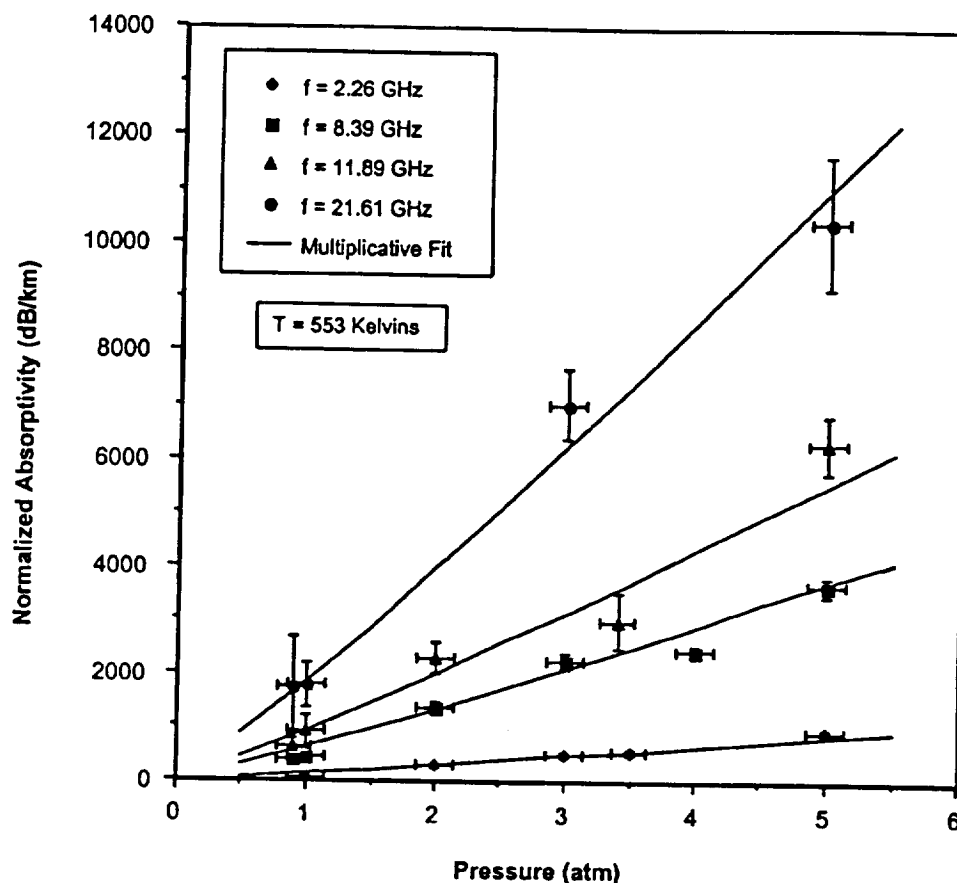


FIG. 6. Laboratory measurements of the microwave absorption normalized to mixing ratio of sulfuric acid vapor as a function of pressure (1 atm = 1.013 bar) in a CO₂ environment.

$$\alpha/q = Ap^B \text{ (over all pressures at one frequency)} \quad (14a)$$

$$\alpha/q = Ap^B f^C \text{ (over all pressures and frequencies),} \quad (14b)$$

where p is the pressure, f is the frequency, A is the amplitude, B is the pressure dependence, and C is the frequency dependence. The parameters A , B , and C are optimized by minimizing the chi-squared function,

$$\chi^2(A, B, C) = \sum_{i=1}^N \frac{1}{\sigma_i^2} ((\alpha/q)_{m_i} - (\alpha/q)_{c_i})^2, \quad (15)$$

where $(\alpha/q)_{m_i}$ is the measured normalized absorptivity, $(\alpha/q)_{c_i}$ is the computed normalized absorptivity from Eqs. (14a) or (14b), σ_i is the error bar of $(\alpha/q)_{m_i}$, and N is the number of measurements.

The σ_i 's for the S- and X-band measurements, though, are on average several factors lower than the σ_i 's for the KU- and K-band measurements due to the higher quality factors (and greater sensitivity) of the large resonator. Therefore, when the chi-squared function for the frequency dependent expression Eq. (14b) is minimized, the

TABLE V
Laboratory Measurements of the Refractivity of Sulfuric Acid Vapor

Experiment number	Measured volume of vaporized solution (cm ³)	Measured refractivity of vaporized solution (N-units)	Normalized refractivity of H ₂ SO ₄ vapor (N-units cm ³ molecule ⁻¹)
1	4.12 ± 0.20	340.64 ± 20.70	(3.230 ± 0.273) × 10 ⁻¹⁶
2	3.18 ± 0.20	245.36 ± 20.71	(2.942 ± 0.343) × 10 ⁻¹⁶
		avg (90% confidence):	(3.086 ± 0.272) × 10 ⁻¹⁶

σ_i 's must all be set to unity. Otherwise, the optimization is too strongly influenced by the S- and X-band measurements resulting in a poor fit at the higher frequencies. This indicates that the frequency dependence of the gaseous H_2SO_4 pressure-broadened absorptivity between the S- and X-band may not be the same as between the X- and KU-band or between the KU- and K-band. The use of a single frequency dependence over the entire microwave spectrum is a limitation of the power law expression Eq. (14b). The σ_i 's are, of course, included in optimizing the parameters A and B in Eq. (14a), which is a frequency independent expression.

We first note that the measurements at X-band were performed at two frequencies, namely 8.39 GHz (Experiment 1) and 8.78 GHz (Experiment 2). Since the Magellan downlink frequency is much closer to the operating frequency in Experiment 1, we perform a frequency correction on the X-band data points from Experiment 2, so that a best fit multiplicative expression may be obtained at 8.39 GHz. The frequency dependence is computed by minimizing the chi-squared function, Eq. (15), for Eq. (14b) over the limited pressure range of the X-band measurements in Experiment 2 (1 to 3 atm). Moreover, only data points at the S-, X-, and KU-band are included, since the normalized absorptivities at these frequencies exhibit a much closer pressure dependence ($p^{1.30}$ to $p^{1.36}$) than those at the K-band ($p^{1.12}$). The resulting frequency correction factor is $f^{1.32}$. The X-band normalized absorptivities from Experiment 2, corrected to 8.39 GHz, are listed in Table IV and plotted in Fig. 3.

Best fit power law expressions are then obtained for all four frequencies. Before considering these expressions, though, the issue of the temperature dependence must be addressed. Both experiments were performed at the same temperature (553 K) to ensure consistency of the data. In order to measure the temperature dependence, consecutive experiments would have to be performed with a temperature differential of at least 20%, which corresponds to about 100 K. If the operating temperature were lowered to 500 K though, very little liquid H_2SO_4 would vaporize and the uncertainties in the mixing ratio would be very large. On the other hand, if the operating temperature were raised to 600 K, the gaskets and hermetic seals in our system would start to break down and the vacuum state, as well as the high pressures, could not be maintained.

The theoretical limits on the temperature dependence of the absorptivity for a non-linear molecule at a line center are $T^{-2.5}$ to $T^{-3.0}$ (Poynter and Pickett, 1985), while the dependence very far from the line center can fall to as low as T^{-4} . The previous S- and X-band multiplicative expressions from Steffes (1985) include a dependence of $T^{-3.0}$, while Fahd (1992) measured a dependence of $T^{-2.9}$ at millimeter wavelengths (94 GHz) for the CO_2 pressure-broadened absorption of sulfuric acid vapor. There has

been extensive research over the past 15 years into the temperature dependence of another trace sulfur-bearing microwave absorber in the Venus atmosphere, namely sulfur dioxide. Janssen and Poynter (1981) computed a multiplicative expression for SO_2 pressure broadened microwave absorption (based on a Van Vleck-Weisskopf formalism) which yielded a temperature dependence of $T^{-3.1}$. This value was consistent with the measurements of Steffes and Eshleman (1981). Suleiman *et al.* (1996) performed lab measurements of the microwave absorption of SO_2 over a wide range of temperatures. The resulting dependencies varied from $T^{-2.8}$ to $T^{-3.0}$. In all cases, the temperature dependence falls within the range of $T^{-3.0 \pm 0.2}$.

Given these rough boundaries, we may be able to estimate temperature dependencies at each of the frequencies based on the variations in the concentrations of the extrapolated microwave lines in the H_2SO_4 line catalog. High concentrations of lines near a frequency would suggest that the majority of contributions to the absorption would be from line centers, indicating a shallower temperature dependence. Low concentrations of lines, on the other hand, would suggest that most of the contributions to the absorption come from the wings of pressure broadened lines, indicating a steeper temperature dependence. The H_2SO_4 line catalog contains 8300 microwave lines (below 30 GHz). The number of lines within a range of ± 500 MHz of each frequency is as follows: 78 lines at 2.26 GHz, 264 lines at 8.39 GHz, 377 lines at 11.89 GHz, and 261 lines at 21.61 GHz. The numbers of lines at X- and K-bands are very close, suggesting that their concentrations represent a mean value over the distribution of microwave lines, and we thus assign these frequencies a temperature dependence of $T^{-3.0 \pm 0.2}$. The number of lines at KU-band is higher and is consequently assigned a slightly shallower dependence of $T^{-2.9 \pm 0.2}$. The number of lines at S-band, on the contrary, is much less than the mean and is therefore assigned a steeper dependence of $T^{-3.2 \pm 0.2}$.

The resulting best fit power law expressions at each frequency for the normalized absorptivity α/q in units of dB km^{-1} are:

$$(\alpha/q)_{2.26 \text{ GHz}} = 103.587 p^{1.356} (553/T)^{3.2 \pm 0.2}, \quad (16a)$$

$$(\alpha/q)_{8.39 \text{ GHz}} = 443.570 p^{1.302} (553/T)^{3.0 \pm 0.2}, \quad (16b)$$

$$(\alpha/q)_{11.89 \text{ GHz}} = 734.735 p^{1.320} (553/T)^{2.9 \pm 0.2}, \quad (16c)$$

$$(\alpha/q)_{21.61 \text{ GHz}} = 1891.486 p^{1.116} (553/T)^{3.0 \pm 0.2}, \quad (16d)$$

where p is the pressure in atm and T is the temperature in K. The corresponding calculated normalized absorptivity for each data point is listed in Table IV. The final column in Table IV, the formalism error, lists the number of standard deviations (σ) that the model deviates from the measured value. Equations (16) are also plotted in

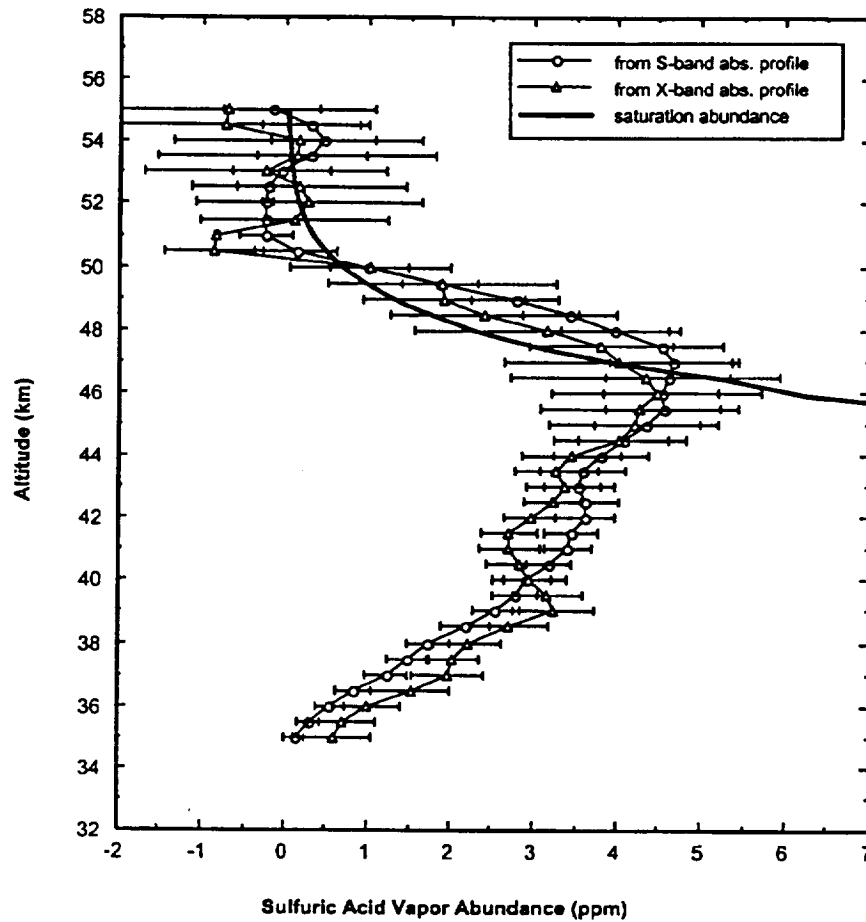


FIG. 7. Abundance profile of gaseous H_2SO_4 below the lower cloud in the Venus atmosphere at a latitude of 67°N , derived from the S- and X-band absorptivity profiles of the Magellan orbit 3212 radio occultation experiment (Jenkins *et al.*, 1994).

Figs. 2–5. Since the pressure dependencies for the S-, X-, and KU-band expressions are close, it would be useful for some applications to have a frequency-dependent best fit expression over all pressures in this limited frequency range. The result is:

$$(\alpha/q)_{\text{S-X-KU}} = 39.471 p^{1.24} f^{1.21} (553/T)^{3.0 \pm 0.2} \text{ dB km}^{-1}, \quad (17)$$

where f is the frequency in GHz. Note that a mean temperature dependence of $T^{-3.0 \pm 0.2}$ has been used. Finally, a frequency-dependent best fit expression is obtained over all pressures and frequencies:

$$(\alpha/q)_{\text{S-X-KU-K}} = 53.601 p^{1.11} f^{1.15} (553/T)^{3.0 \pm 0.2} \text{ dB km}^{-1}. \quad (18)$$

As expected, the pressure dependence of Eq. (18) is strongly influenced by the K-band data points. This expression, plotted in Fig. 6, would therefore be most useful for application within the vicinity of the K-band.

8. APPLICATION TO VENUS RADIO OCCULTATION RESULTS

8.1. Summary of Absorptivity Profiles

We now apply the new expressions for the S-band and X-band microwave opacity of sulfuric acid vapor to the Magellan and Mariner 10 radio occultation results. Complete descriptions of the design and performance of the Magellan and Mariner 10 radio occultation experiments are given respectively in Steffes *et al.* (1994) and Howard *et al.* (1974). Both refractivity profiles and absorptivity profiles are derived from occultation experiments. Density, pressure, and temperature profiles are inferred from the refractivity profiles using the known number density normalized refractivities of CO_2 and N_2 (Essen and Froome, 1951) for an atmospheric composition of 96.5% CO_2 and 3.5% N_2 , which is assumed to be in hydrostatic equilibrium.

Absorptivity profiles at both S- and X-band were obtained from radio occultation experiments in October of

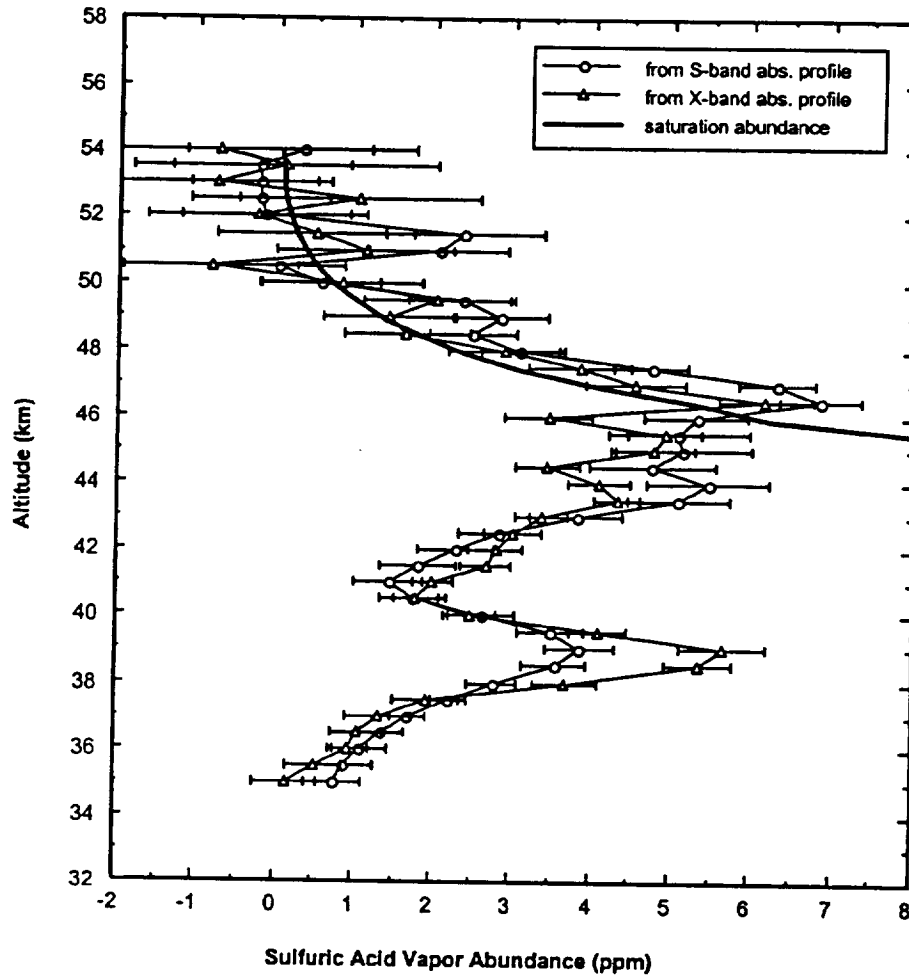


FIG. 8. Abundance profile of gaseous H_2SO_4 below the lower cloud in the Venus atmosphere at a latitude of 67°N , derived from the S- and X-band absorptivity profiles of the Magellan orbit 3213 radio occultation experiment (Jenkins *et al.*, 1994).

1991 during three consecutive orbits (3212, 3213, and 3214) of the Magellan spacecraft (Jenkins *et al.*, 1994). The latitude of the region measured by each occultation was 67°N . Note that the Venus atmosphere has a 5-day rotation period, and thus the atmosphere rotates about 10° between each 3.26-h orbit of the spacecraft. Likewise, S-band absorptivity profiles were obtained from occultation experiments in December of 1992 during two consecutive orbits (6369 and 6370) of the spacecraft (Hinson and Jenkins, 1995). The latitude measured by these occultations was 88°S . Data from a series of experiments performed in the summer of 1994 at latitudes 49°N , 74°N , 82°N , 62°S , and 87°S are currently being processed (Jenkins, personal communication). Finally, an equatorial absorptivity profile at S-band (2.295 GHz) was attained from the Mariner 10 radio occultation experiment (Lipa and Tyler, 1979). This profile is both consistent with and more complete than the equatorial S-band absorptivity profiles obtained from the Pioneer-Venus orbiter (Jenkins and Steffes, 1991).

8.2. Abundance Profiles of Gaseous H_2SO_4

To compute profiles of gaseous sulfuric acid abundance in the Venus atmosphere, the pressure, temperature, and absorptivity profiles from the radio occultation results are first partitioned and matched to discrete intervals. Profiles from Magellan orbits 3212, 3213, and 3214 and the Mariner 10 equatorial profile are partitioned to 0.5 km intervals. Magellan orbits 6369 and 6370 are partitioned to 0.2 km intervals. Next, the absorption due to CO_2 and N_2 collisionally induced dipole moments is removed from the absorptivity profiles using the expression from Ho *et al.* (1966):

$$\alpha_{\text{CO}_2} = 1.15 \times 10^8 (q_{\text{CO}_2}^2 + 0.25 q_{\text{CO}_2} q_{\text{N}_2} + 0.0054 q_{\text{N}_2}^2) f^2 p^2 T^{-5}, \quad (19)$$

where f is the frequency in GHz, p is the pressure in atm, T is the temperature in K, q is the number mixing ratio

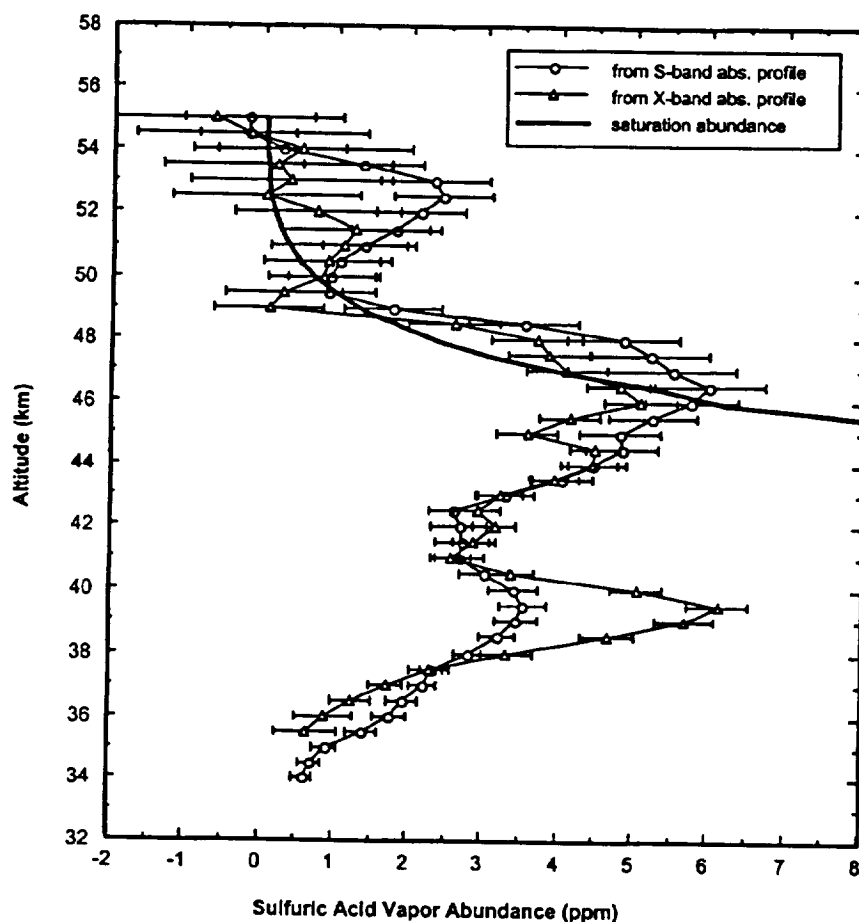


FIG. 9. Abundance profile of gaseous H_2SO_4 below the lower cloud in the Venus atmosphere at a latitude of 67°N , derived from the S- and X-band absorptivity profiles of the Magellan orbit 3214 radio occultation experiment (Jenkins *et al.*, 1994).

(0.965 for CO_2 and 0.035 for N_2), and α is the absorption in units of dB km^{-1} .

The remaining absorption is principally due to H_2SO_4 vapor. Equations (16a) and (16b) are then applied independently to the residual S- and X-band absorptivity profiles to yield gaseous H_2SO_4 abundance profiles. A small frequency correction factor is applied ($f^{1.21}$ from Eq. (17)) to precisely match the 2.26 GHz and 8.39 GHz expressions to the Mariner 10 and Magellan S- and X-band downlink frequencies (2.30 GHz and 8.43 GHz). The abundance profiles derived from both the S- and X-band absorptivity profiles for orbits 3212, 3213, and 3214 are shown in Figs. 7–9. The abundance profiles derived from the S-band absorptivity profiles for Magellan orbits 6369 and 6370 and the Mariner 10 equatorial profile are shown respectively in Figs. 10, 11, and 12.

The plotted 1- σ error bars on the gaseous H_2SO_4 abundances represent only the error bars in the measured absorptivities from the radio occultation experiments. The primary uncertainty in our expressions for the S- and X-

band H_2SO_4 vapor opacity is the temperature dependence. This adds 5% uncertainty to the abundance error bars in the 38 to 40 km region, increasing to an additional 8% uncertainty at 48 km. Also plotted in Figs. 7–12 is the saturation abundance of H_2SO_4 . Our expression for the H_2SO_4 saturation vapor pressure (Kolodner and Steffes, 1994) is a slight modification of the Clausius–Clapeyron model developed by Ayers *et al.* (1980), which incorporates the temperature dependence of the enthalpy of vaporization in its development. This expression is consistent with both the measured H_2SO_4 saturation vapor pressures by Ayers *et al.* (1980) in the 338 to 445 K temperature range as well as the calculated vapor pressures by Vermeulen (1984) at temperatures of 500 K and higher. At the cloud base, we assume a sulfuric acid concentration of 98.2% by weight, as modeled by Krasnopolsky and Pollack (1994).

8.3. Comparative Analysis

The abundances of gaseous H_2SO_4 range from 10–14 ppm below a 48–48.5 km altitude cloud layer at the equa-

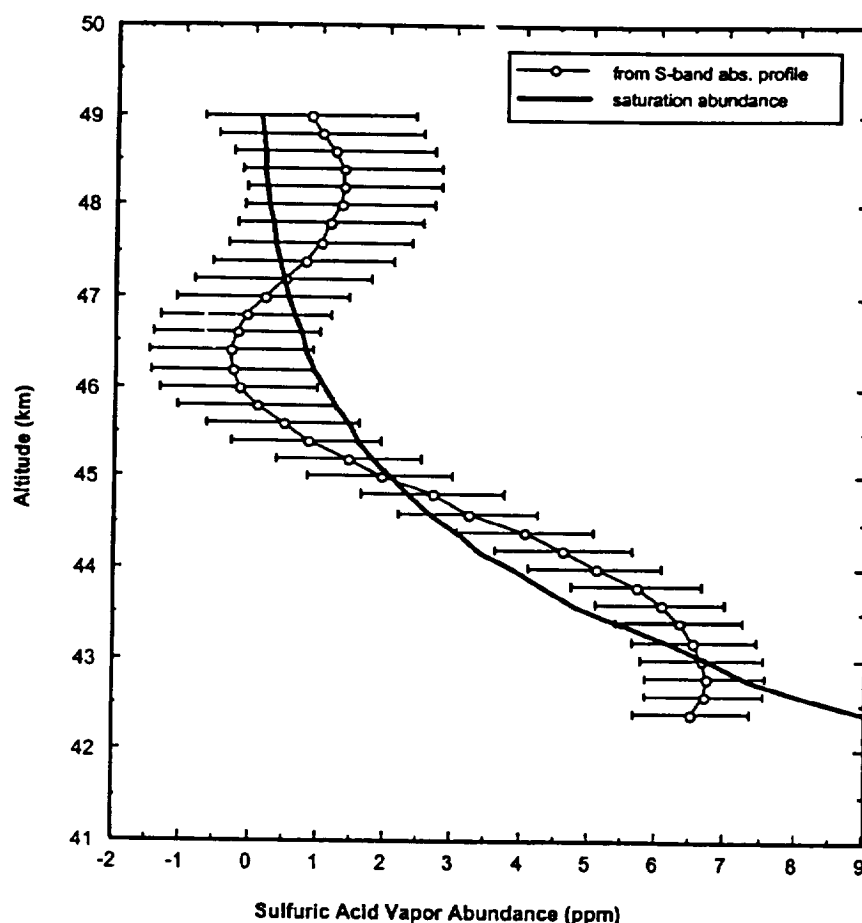


FIG. 10. Abundance profile of gaseous H_2SO_4 below the lower cloud in the Venus atmosphere at a latitude of 88°S , derived from the S-band absorptivity profile of the Magellan orbit 6369 radio occultation experiment (Hinson and Jenkins, 1995).

tor, to 3–7 ppm below a 47–47.5 km cloud layer base at 67°N , finally to 6–10 ppm below a 43.5–44 km cloud layer base at 88°S . The magnitudes of these abundances are in complete agreement with the H_2SO_4 vapor profiles computed by Krasnopolsky and Pollack (1994). The thermal dissociation of gaseous H_2SO_4 into SO_3 and H_2O is evident in the profiles of Magellan orbits 3212, 3213, and 3214 as well in the Mariner 10 profile. The profiles from Magellan orbits 6369 and 6370 do not probe deep enough to see the H_2SO_4 vapor abundance fall back to zero.

The gaseous H_2SO_4 abundance profiles derived independently from the S- and X-band absorptivity profiles of the October 1991 Magellan radio occultation experiments are, for the first time, consistent with one another. The exception, of course, is the 38–40 km region of orbits 3213 and 3214, where the X-band abundances are 2 to 3 ppm higher than the S-band abundances. The fact that we do not see this feature in orbit 3212 suggests that the Venus atmosphere rotated between orbits 3212 and 3213 into a region containing an elevated amount of another trace constituent

which absorbs much more strongly at X-band than at S-band. The obvious candidate for this other constituent is sulfur dioxide (SO_2). The H_2SO_4 vapor abundance profiles from orbits 3213 and 3214 peak 1 to 2 ppm higher than the profile from orbit 3212. This suggests an elevated amount of the gaseous H_2SO_4 dissociate SO_3 in the 38–40 km region of these orbits, which will react with carbon monoxide (CO) to produce additional CO_2 and an increase in the abundance of SO_2 (Krasnopolsky and Pollack, 1994). The microwave opacity formalism for pressure broadened SO_2 has been developed by Suleiman *et al.* (1996). While typical amounts of SO_2 (75 to 150 ppm) cannot fully resolve the discrepancy between the S- and X-band abundance profiles, its addition helps to converge the S- and X-band H_2SO_4 vapor abundances in this region due to the frequency squared dependence of its microwave absorption.

The vertical structure of the gaseous H_2SO_4 abundance profile at the equator (Fig. 12) is quite different than the vertical structure of the abundance profiles near the poles (Figs. 7–9). Preceding the formation of the cloud near 48

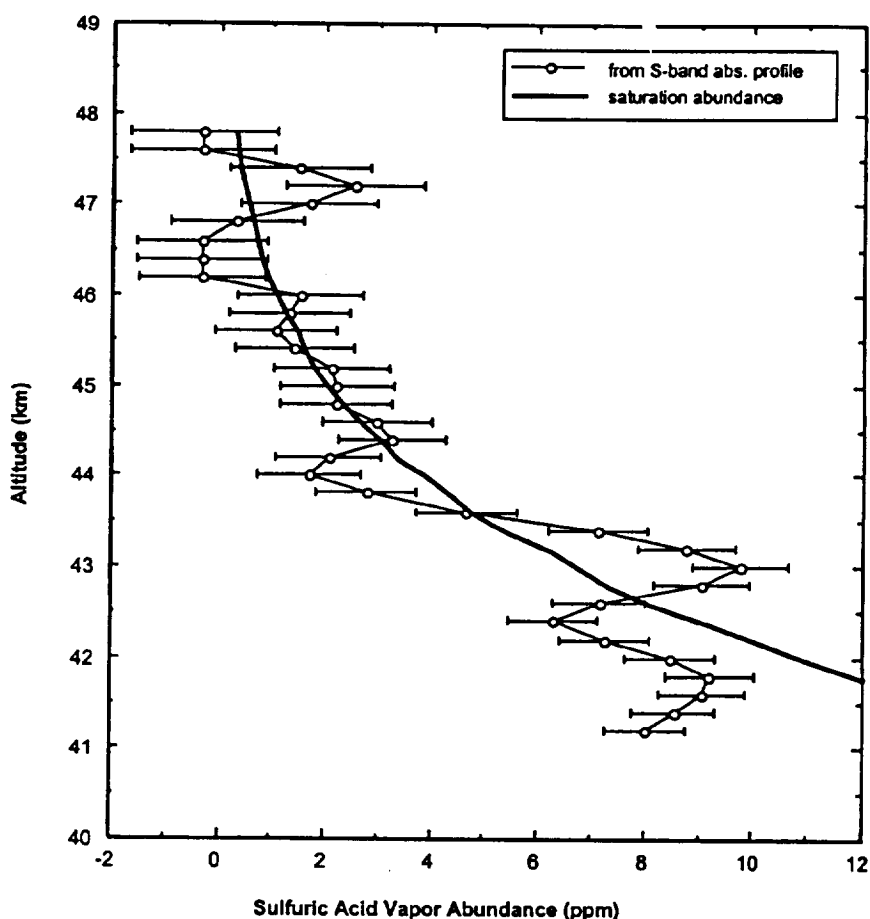


FIG. 11. Abundance profile of gaseous H_2SO_4 below the lower cloud in the Venus atmosphere at a latitude of 88°S , derived from the S-band absorptivity profile of the Magellan orbit 6370 radio occultation experiment (Hinson and Jenkins, 1995).

km at the equator, a very high and sharp peak in H_2SO_4 vapor abundance (14 ppm) occurs near 45 km. Below this altitude, the abundance profile drops off very suddenly as gaseous H_2SO_4 dissociates into H_2O and SO_3 . In contrast, the Magellan orbit 3212 abundance profile, for example, does not peak as high as the Mariner 10 profile directly below the cloud (5 ppm), but remains undissociated over a greater distance below the cloud. The thermal dissociation of H_2SO_4 vapor does not commence until an altitude of 40 km is reached.

A second difference between equatorial and polar abundance profiles is their degree of supersaturation. The Mariner 10 profile (Fig. 12) agrees quite well with the saturation abundance profile. In fact, most Venus cloud models show that supersaturation should be minimal at the lower cloud base (LCB), since sulfuric acid particles at the LCB do not grow rapidly in number but merely nucleate upon some preexisting particles (Esposito *et al.*, 1983). The Magellan gaseous H_2SO_4 abundance profiles (Figs. 7–11) do exhibit a moderate amount of supersaturation. It was also pointed

out by Esposito *et al.* (1983), though, that large supersaturations could exist in regions of the atmosphere affected by an adiabatic cooling mechanism such as strong downwelling vertical motion. The differences between the equatorial and polar gaseous H_2SO_4 abundance profiles indicate the presence of strong zonal climatic variability near the cloud base in the Venus atmosphere.

8.4. Refractivity of H_2SO_4 Vapor

Finally, our new value for the number density normalized refractivity of gaseous H_2SO_4 (Table V) can be used in conjunction with the derived H_2SO_4 vapor abundances to determine what percent contribution to the overall refractivity directly below the cloud layer is due to H_2SO_4 vapor. The total number density at an altitude of 45 km and a latitude of 67°N in the Venus atmosphere is 3.5×10^{19} molecule cm^{-3} . The upper limit on the abundance of gaseous H_2SO_4 in this region is 10 ppm. The resulting refractivity of H_2SO_4 vapor, using our result from Table V, is 0.108 N-units (upper limit). The total refractivity

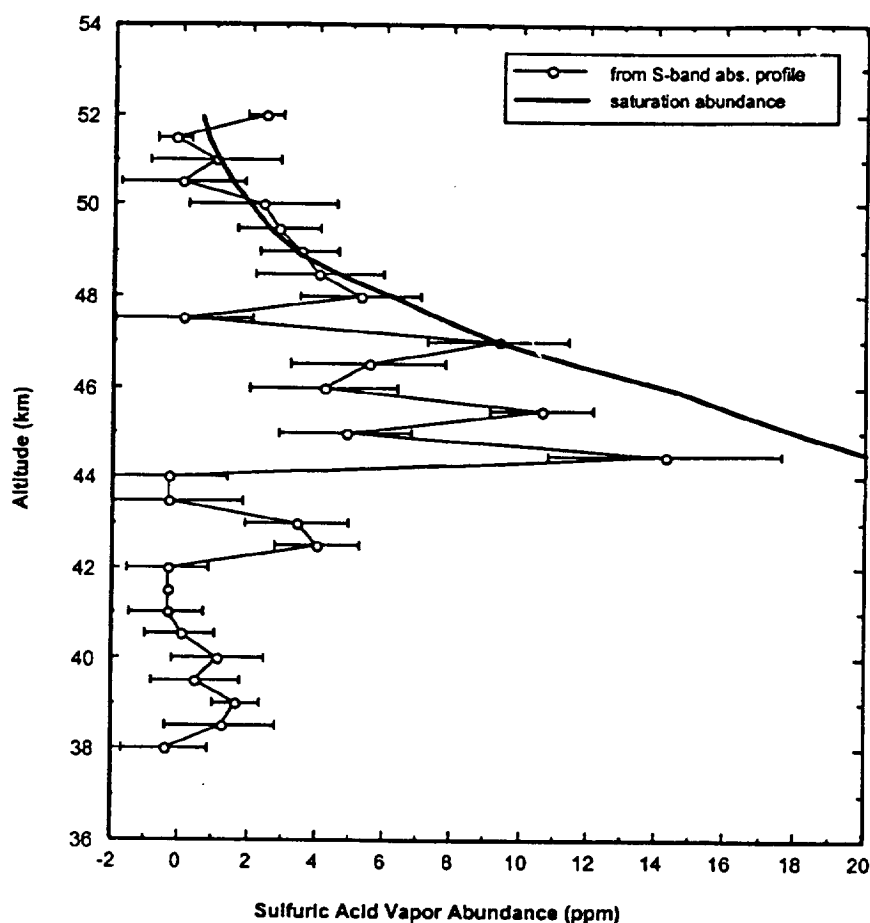


FIG. 12. Abundance profile of gaseous H_2SO_4 below the lower cloud in the Venus atmosphere at the equator, derived from the S-band absorptivity profile of the Mariner 10 radio occultation experiment (Lipa and Tyler, 1979).

measured at this altitude during the 1991 Magellan radio occultation experiments is 633 N-units, yielding a net contribution from H_2SO_4 vapor of 0.017% (upper limit). This contribution is barely detectable and is roughly an order of magnitude below the measured variations in refractivity from orbit to orbit during the 1991 radio occultation experiments (Jenkins *et al.*, 1994). Our results, though, suggest a strong correlation between spatial variations in gaseous H_2SO_4 abundance and the local and global dynamical processes taking place in the Venus atmosphere.

9. CONCLUSIONS AND SUGGESTIONS FOR FUTURE WORK

The first portion of this work described the completion of new laboratory measurements of the microwave absorption of sulfuric acid vapor under simulated Venus conditions. Several improvements have been made on the previous experimental setup and procedure (Steffes, 1985 and 1986) to reduce the error bars of the experimental data.

First, the use of cylindrical resonators plated with gold as opposed to silver has significantly reduced the amount of chemical decomposition of the sulfuric acid vapor and thus greatly increased our ability to accurately compute the H_2SO_4 number mixing ratio. Second, a significant reduction in the error bars of the absorptivity measurements has been achieved due to our ability to account for changes in the dielectric properties of the resonators when a lossy gaseous mixture is introduced into them. Last, a data acquisition system has been added (DeBoer and Steffes, 1996) which allowed the data to be recorded and averaged in a more systematic and reliable fashion.

The measured absorptivities normalized to mixing ratio are several factors higher than those measured by Steffes (1985, 1986). This is principally due to the overstatement of the mixing ratio due to unaccounted for chemical reactions between the acid and the silver plating of the resonators in the previous experiments. New power law multiplicative expressions for the pressure broadened absorption of gaseous H_2SO_4 have been produced at frequencies 2.26, 8.39,

11.89, and 21.61 GHz. In addition, frequency dependent multiplicative expressions have been best fit to the data. Finally, the refractivity of H_2SO_4 vapor normalized to number mixing ratio has been accurately measured for the first time.

The S- and X-band power law expressions for the opacity of H_2SO_4 vapor have been applied to the Magellan and Mariner 10 radio occultation experiments to yield specific and self-consistent abundance profiles of gaseous H_2SO_4 below the main cloud layer in the Venus atmosphere. The new abundance profiles, which closely follow the H_2SO_4 saturation abundance, will be useful in future thermochemical and microphysical modeling of the Venus atmosphere and clouds. The observed variations in the abundances of gaseous H_2SO_4 vertically, longitudinally (orbit to orbit), and latitudinally are statistically significant and signify the presence of local and global dynamical processes in the Venus atmosphere.

New dynamical features are sure to be detected from the 10 Magellan radio occultation experiments performed between June and August of 1994 at a variety of latitudes in the Venus atmosphere. The results of these experiments will be presented in an upcoming paper. Last, a dual-frequency microwave observation of Venus has been performed at 1.3 and 2 cm using the Very Large Array (VLA). A complete description of the observation and its interpretation will also be presented in upcoming papers. Significant darkening was observed in the polar regions (>25 K), which could suggest elevated amounts of gaseous sulfuric acid and sulfur dioxide. Our derived equatorial and polar gaseous H_2SO_4 abundance profiles are being used in a newly developed microwave radiative transfer model to invert the received VLA radiances and infer spatial variations in gaseous H_2SO_4 abundance across the disk of the planet. These VLA maps coupled with the new abundance profiles of H_2SO_4 vapor from the Magellan and Mariner 10 radio occultation experiments will give greater insight into the global dynamical features and climatology of the Venus atmosphere.

ACKNOWLEDGMENTS

The authors first thank Dr. David R. DeBoer of the Georgia Institute of Technology for providing the data acquisition software used in these experiments. We also thank Kings Electronics Corporation for donating the high-temperature N-type hermetically sealed bulkhead feedthroughs, Belden Corporation for donating the high-temperature coaxial cable, Dupont Corporation for donating the high-concentration sulfuric acid solution, and Mr. Ben Johnson of Techtrix, who went beyond the call of duty to ensure that we had the best gold-plating possible. Finally, we acknowledge Dr. Shady H. Suleiman and Dr. Scott A. Borgsmiller from the Georgia Institute of Technology for their invaluable assistance in assembling the experimental setup. This work was supported by the Planetary Atmospheres Program of the National Aeronautics and Space Administration under Grants NAGW-533 and NAG5-4190.

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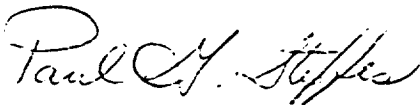
September 23, 1997

Dr. Barry G. Clark
Dr. W. Miller Goss
National Radio Astronomy Observatory - VLA
1003 Lopezville Rd
Socorro, NM 87801

Dear Drs. Clark and Goss:

Enclosed is a "Target of Opportunity" proposal for use of the VLA for a 12 hour observation of Venus in December 1997 in conjunction with a simultaneous infrared observation to be conducted at the Anglo-Australian Telescope (AAT). Since the opportunity for simultaneous IR/microwave mapping of Venus will not present itself again until mid-2001, we would deeply appreciate the Time Allocations Committee's consideration on our behalf.

Sincerely,



Paul G. Steffes
Professor



VLA OBSERVING APPLICATION

DEADLINES: 1st of Feb., June., Oct. for next configuration following review

INSTRUCTIONS: Each numbered item must have an entry or N/A

SEND TO: Director NRAO, 520 Edgemont Rd., Charlottesville, VA 22903-2475

A

rcvd:

(1) Date Prepared: September 23, 1997

(2) Title of Proposal: Simultaneous microwave/NIR imaging of Venus

(3) AUTHORS	INSTITUTION	Who Will Come To The VLA?	For Grad Students Only	
			Observations For Ph.D. Thesis?	Anticipated Ph.D. Year
Paul G. Steffes	Georgia Institute of Technology	X		
Bryan J. Butler	National Radio Astronomy Observatory	X		
Jon M. Jenkins	NASA-Ames Research Center			

(4) Related VLA previous proposal number(s): AS 578

(5) Contact author Paul G. Steffes

for scheduling: School of Elec. & Computer Engr.

address: Georgia Institute of Technology
Atlanta, GA 30332 0250

(6) Telephone: 404 894 3128

Telex:

Internet: ps11@prism.gatech.edu

Other E Mail:

Telefax: 404 894 5935

(7) Scientific Category: ☐ astrometry, geodesy & techniques, ☐ solar, ☐ propagation, ☒ planetary, ☐ stellar, ☐ pulsar, ☐ ISM, ☐ galactic center, ☐ galactic structure & dynamics (HI), ☐ normal galaxies, ☐ active galaxies, ☐ cosmology

(8) Configurations (one per column) (A, B, C, D, BnA, CnB, DnC, Any)	D				
(9) Wavelength(s) (400, 90, 20, 18, 6, 3.5, 2, 1.3, 0.7 cm)	1.3 and 2cm				
(10) Time requested (hours)	12				

(11) Type of observation: ☒ mapping, ☐ point source, ☐ monitor, ☒ continuum, ☐ lin poln, ☐ circ poln, ☐ solar, ☐ VLBI, (check all that apply) ☐ spectroscopy, ☐ multichannel continuum, ☐ phased array, ☐ pulsar, ☐ high-time resolution ☐ other _____

(12) ABSTRACT (Do not write outside this space. Please type.)

A successful mapping of Venus at 1.3cm and 2 cm was conducted in April 1996. Comparison of the microwave maps with 2 micron infrared maps indicates an anti-correlation between the IR maps and the microwave maps, which may indicate Hadley-cell type circulation in the Venus atmosphere. December 1997 will be the only opportunity until Summer 2001 in which it will be possible to simultaneously image Venus in the near-IR and the VLA. Thus a compelling "Target of Opportunity" exists.

NRAO use only

- (13) Observer present for observations? ☒ Yes ☐ No Data reduction at? ☐ Home ☒ AOC or CV (2 weeks notice)
- (14) Help required: ☒ None ☐ Consultation ☐ Friend (extensive help)

(15) Spectroscopy Only:

	line 1	line 2	line 3	line 4
Transition (HI, OH, etc.)	_____	_____	_____	_____
Rest Frequency (MHz)	_____	_____	_____	_____
Velocity (km/s)	_____	_____	_____	_____
Observing frequency (MHz)	_____	_____	_____	_____
Correlator mode	_____	_____	_____	_____
IF bandwidth(s) (MHz)	_____	_____	_____	_____
Hanning smoothing (y/n)	_____	_____	_____	_____
Number of channels per IF	_____	_____	_____	_____
Frequency Resolution (kHz/channel)	_____	_____	_____	_____
Rms noise (mJy/bm, nat. weight., 1 hr)	_____	_____	_____	_____
Rms noise (K, nat. weight., 1 hr)	_____	_____	_____	_____

- (16) Number of sources 1 (If more than 10 please attach list. If more than 30 give only selection criteria and LST range(s).)

(17) NAME	Epoch: 1950 <input type="radio"/> 2000 <input type="radio"/> RA Dec hhmm ± xx.x°	Config.	Band (cm)	Band- width (MHz)	Total Flux		Largest angular size	Required rms (mJy/bm)	Time requested (hours)
					line (Jy)*	cont. (Jy)			
Venus	20 10 -22.0	D	1.3	50	N/A	202	43"	0.072	2
		D	2	50	N/A	92	43"	0.042	

*this should be the total flux at the peak of the line

Notes to the table (if any):

- (18) Restrictions to elevation (other than hardware limits) or IIA range (give reason): None
- (19) Preferred range of dates for scheduling (give reason): December 10-15, 1997 (See attachment)
- (20) Dates which are not acceptable: All other times.
- (21) Special hardware, software, or operating requirements: N/A
- (22) Please attach a self-contained Scientific Justification not in excess of 1000 words. (Preprints or reprints will be IGNORED!)
- Please include the full addresses (postal and e-mail) for first-time users or for those that have moved (if not contact author).
- When your proposal is scheduled, the contents of the cover sheets become public information (Any supporting pages are for refereeing only).

v3.0 6/94

MEMORANDUM

TO: VLA Time Allocation Committee

FROM: Prof. Paul G. Steffes (Georgia Institute of Technology, School of Electrical and Computer Engineering, Atlanta, GA 30332-0250 (404) 894-3128, e-mail: ps11@prism.gatech.edu)

Dr. Jon M. Jenkins (NASA Ames Research Center, M/S 245-3, Moffett Field, CA 94035-1000 (650) 604-6524, e-mail: jjenkins@mail.arc.nasa.gov)

Dr. Bryan J. Butler (NRAO, P.O. Box O, Socorro, NM 87801 (510) 835-7261, e-mail: bbutler@aoc.nrao.edu)

SUBJECT: Target of Opportunity, December 1997

In April 1996, Suleiman, Kolodner, Butler and Steffes conducted a highly successful mapping effort of Venus from the VLA at 1.3 and 2.0 cm. The results were especially striking in that pronounced darkening in the polar regions (relative to a reference atmosphere) was readily visible in the maps, which correlated with enhanced microwave absorption measured in the polar regions by Magellan radio occultation experiments (Jenkins, ICARUS 110, 79-94, June 1994). These results are currently being prepared for publication and were presented at the 1997 AAS/DPS Meeting (B.A.A.S. v.19, 1042-1043, 1997). At the AAS/DPS Meeting, the two groups of astronomers who had conducted IR observations noted that the radio maps appeared anti-correlated with the IR maps, which is consistent with the idea that cloud densities (which predominate IR opacities) are reduced in the polar zones, but the abundance of vaporization products (gaseous H₂SO₄ and SO₂, which predominate the non-CO₂ microwave opacity) are elevated in the polar zones. Such results would be strong indicators of Hadley-cell type circulation which has been thought to be a potential driver of the enigmatic superrotation of the Venus atmosphere.

Unfortunately, since past microwave and IR observations were taken at different epochs, the results to date have been only suggestive of this theory. It was agreed that simultaneous or near-simultaneous observation of Venus at 1.3 cm, 2 cm, and in the near IR would be required to provide more substantive proof of this theory. On December 10-15, 1997, a near IR Venus mapping campaign will be conducted from the 3.9-meter Anglo-Australian Telescope (AAT), using the Max Planck 3D imaging spectrometer. Since on those dates Venus will be approximately 40 days short of inferior conjunction, its large size will make moderate resolution mapping possible with the VLA in the D-configuration. (During this period, Venus will have a diameter of approximately 41.6 arcseconds, which will subtend 10.6 beams at U-Band, and 14.8 beams at K-Band.) Since the next opportunity to observe Venus with the VLA occurs in the next B-configuration when Venus is 10.1 arcseconds wide and only 13 degrees separate from the sun, it would not be possible to observe in the infrared. The next opportunity to observe Venus when its size and elongation are large enough so that it can be imaged by the IR instruments, AND so that the VLA will be

operating in the C- or D- configuration will not occur until Summer 2001. Thus, this will be the only opportunity in the near future to conduct the necessary joint observation.

As a result, we respectfully request observation time in the 10-15 December time period, as a "Target of Opportunity" observation. We request one 12-hour track to observe Venus in the time period December 10-15.

Dr. Victoria Meadows
MS 183-900
Jet Propulsion Laboratory
4800 Oak Grove Drive
Pasadena CA 91109-8099
(818) 354 0528
vsm@hesperos.jpl.nasa.gov

September 19, 1997

Dr. Barry G. Clark and Dr. W. Miller Goss
NRAO-VLA Time Allocation Committee
P.O. Box 0
Socorro, NM 87801

Dear Sirs

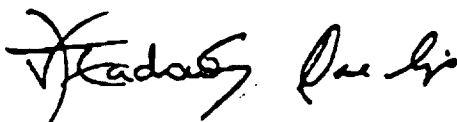
The following is written in support of the application submitted by Prof. Paul Steffes for coordinated near-IR and VLA observations of Venus prior to the 16 January 1998 inferior conjunction.

The planet Venus is completely shrouded by an optically-thick cloud layer that is composed of concentrated sulfuric acid (H_2SO_4) aerosols. These clouds, combined with the massive 90-bar CO_2 atmosphere, support an effective greenhouse mechanism which produces surface temperatures near 730K. The clouds also modulate atmospheric trace constituent abundances by providing both a local reservoir of water vapor and sulfur species, and providing surfaces on which heterogeneous (gas-solid) chemical reactions can occur. Many of these trace gases, such as SO_2 and H_2O , play key roles in the atmospheric chemistry and greenhouse mechanism, and their abundances can serve as tracers of large-scale circulations in the Venus atmosphere. To better understand the cloud-level physical, chemical and dynamical processes, improved observations of the abundances and spatial distributions of the cloud and sub-cloud region (20-45km) are needed. However, the planet-wide Venus clouds are highly reflective, and preclude observations of the sub-cloud region at visible wavelengths. For ground-based observers, near-infrared and radio observations are required to probe below the top of the Venus cloud deck.

We have recently been awarded observing time on the 3.9m Anglo-Australian Telescope (AAT) to use the new Max Planck 3D imaging spectrometer for near-infrared observations of Venus on the evenings of 10-15 December, 1997. Using this instrument we will obtain spatially resolved maps of sub-cloud water abundance (in the altitude range 25-45km) and variations in the cloud H_2SO_4 aerosol opacity in the range 45-48km altitude, but will not be sensitive to H_2SO_4 or SO_2 vapor abundance, the remaining key components of the cloud dissociation process. However, maps of these remaining constituents can be obtained with VLA observations at 1.3 and 2cm, which have weighting functions that probe comparable

altitude regions. We therefore feel it would be extremely beneficial scientifically to take advantage of a rare opportunity to coordinate near-infrared and radio observations of the Venus sub-cloud atmosphere. With coordinated observations on one or more of our scheduled near-infrared observing dates, we will be able to obtain near-simultaneous spatially resolved maps of sub-cloud H_2O , SO_2 and H_2SO_4 vapor and aerosols. These maps will be used to search for correlations in gas abundances with aerosol concentration that characterize cloud subsidence, and cloud formation and dissociation processes. The derived abundances would also provide more stringent constraints on cloud chemistry and the Venus greenhouse mechanism.

Sincerely,

A handwritten signature in black ink, appearing to read "Victoria Meadows" followed by a flourish and then "David Crisp".

Dr. Victoria Meadows

Dr. David Crisp

THE GEORGIA TECH HIGH SENSITIVITY MICROWAVE MEASUREMENT SYSTEM

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Abstract. As observations and models of the planets become increasingly more accurate and sophisticated, the need for highly accurate laboratory measurements of the microwave properties of the component gases present in their atmospheres become ever more critical. This paper describes the system that has been developed at Georgia Tech to make these measurements at wavelengths ranging from 13.3 cm to 1.38 cm with a sensitivity of 0.05 dB/km at the longest wavelength and 0.6 dB/km at the shortest wavelength.

1. Introduction

One of the greatest hindrances in modeling and fitting synthetic spectra of the planets is the lack of realistic models of the absorption profiles of the atmospheric components under planetary conditions (see e.g., de Pater and Mitchell, 1994). A program has been underway at the Georgia Institute of Technology to measure the absorptive and refractive properties of these components as a function of temperature, pressure, composition and frequency under conditions similar to those on Venus (Steffes, 1985; Fahd and Steffes, 1991; Fahd and Steffes, 1992) and the outer planets (Steffes and Jenkins, 1987; Jenkins and Steffes, 1988; Joiner *et al.*, 1989; DeBoer and Steffes, 1994). As the thermochemical and radiative transfer models of the planets become more sensitive and sophisticated the need for increasingly accurate measurements is apparent. This paper reviews the process of determining the refractive index and absorption coefficients of low-loss gases and describes the high sensitivity microwave measurement system used at Georgia Tech to measure these values, including a full discussion of the new computer interface and analysis system which greatly enhances the sensitivity and reliability of the results.

2. System Overview

2.1. DATA ANALYSIS

The Georgia Tech system (see Figure 1) uses the changes in the quality factor (Q) and center frequency of a resonator with a test gas present to measure absorptivity and refractivity. These changes are monitored using a swept signal source and a

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spectrum analyzer whose output is read into a personal computer for display and analysis. The refractivity of the test gas is measured by noting the change in the center frequency of the resonance with the test gas present relative to the center frequency in the presence of a vacuum. The refractivity of the gas (defined as $N_R = 10^6(n - 1)$, where n is the refractive index of the gas) is given by

$$N_R = 10^6 \frac{f_v - f_g}{f_g} \quad (1)$$

where f_v and f_g are the center frequencies of the resonance with a vacuum and test gas present, respectively (see e.g., Tyler and Howard, 1969).

To determine the absorptivity of the test gas, the quality factor (Q) of the resonance is then measured a number of times with the test gas present to develop statistics of the measurement. The expression for the quality factor is given by

$$Q = \frac{f_g}{\Delta f} \quad (2)$$

where, again, f_g is the center frequency and Δf is the full-width half maximum power (FWHM) bandwidth of the resonance. The test gas is then vented and a reference, lossless gas is added such that the change in center frequency from a vacuum is the same for this reference gas as it was for the test gas. This is to compensate for the effects of dielectric loading; that is, when Equation 3 (to be introduced shortly) is not strictly valid due to changes in the real part of the refractive index (see e.g., DeBoer and Steffes, 1994 for a brief mathematical explanation and Spilker (1990) for a physical discussion of dielectric loading). The quality factor of the resonance is then measured with the reference gas and the absorptivity of the test gas is computed via

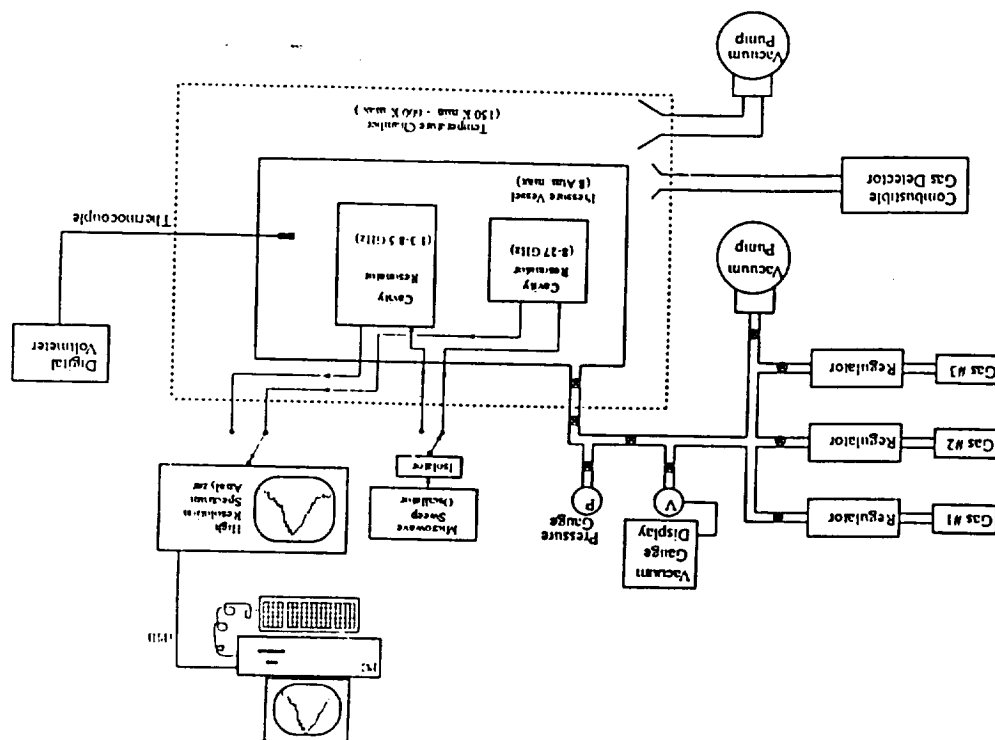
$$\alpha = 4.343 \frac{2\pi}{\lambda} \left(\frac{1 - \sqrt{t_r}}{Q_g} - \frac{1 - \sqrt{t_r}}{Q_r} \right) \text{ dB/m} \quad (3)$$

where λ is the wavelength in meters at the center of the resonance with the test gas present and Q and t refer to the measured quality factor and transmissivity (the portion of the power that is transmitted through the resonator at the resonance center) of the resonator with the test gas (subscript g) and reference gas (subscript r) present.

Errors in this calculation are due to the finite sensitivity and accuracy of the spectrum analyzer (which are minimized by proper selections of the spectrum analyzer parameters such as resolution bandwidth and analyzer span) and by electrical, stochastic noise (which is minimized by maximizing the signal-to-noise ratio and by taking and collecting many samples). For a more complete discussion of the techniques for data analysis, as well as a complete description of the measurement error and error determination, see DeBoer and Steffes (1994).

Errors in measuring the properties of the environment in which the absorptivity measurements took place (e.g., temperature, pressure . . .) do not affect the application or analysis of the above equation, i.e., the measured absorptivity is the value

Figure 1. Block diagram of the Georgia Tech planetary atmospheric simulator and data collection/analysis sub-systems.



for that environment. The environmental measurement errors manifest themselves when the measured absorption is fit to some sort of formalism to mathematically model the absorption.

Absorption is typically calculated by summing over the contributions from that molecule's set of absorption lines, unless the frequency of interest lies in the so-called continuum and a power law formalism can be fit. For a set of absorption lines, the absorption may be calculated via

$$\alpha = \sum_i n(P, T) S_i(T) F_i(\nu, \nu_i, P, T, \dots) \quad (4)$$

where $n(P, T)$ is the number density of the absorber, $S_i(T)$ is the line intensity, and $F_i(\nu, \nu_i, P, T, \dots)$ is the lineshape function as a function of frequency, line center, pressure, temperature, etc. See, for example, Townes and Schawlow (1955). Obviously, the temperature and pressure of the sample must be known to apply this equation. To first order (and assuming an ideal gas), variation in one term of this sum can be written

$$\delta\alpha = \chi J(T) P \frac{\partial F}{\partial \nu} \delta\nu + \chi J(T) \left[F + P \frac{\partial F}{\partial P} \right] \delta P \\ + \chi J(T) P \left[\frac{hcE}{kT^2} F - \frac{3}{T} F + \frac{\partial F}{\partial T} \right] \delta T + J(T) P F \delta\chi \quad (5)$$

where

$$J(T) = \frac{A}{T_0} \left(\frac{T_0}{T} \right)^3 e^{-hcE/kT} \quad (6)$$

and A is a constant and χ is the mixing ratio of the absorbing gas. (Note, this equation also assumes that $\partial F / \partial \chi \approx 0$; i.e., the linear dependence of α on χ dominates).

The coefficients of the various terms show the magnitudes of the effects resulting from the uncertainty in measuring that term. Figure 2 plots those coefficients for a Van Vleck-Weisskopf lineshape assuming parameters similar to the 168 GHz absorption line of H_2S and an H_2S mixing ratio (χ) of 10%.

2.2. LABORATORY CONFIGURATION

The general characteristics of this type of measurement system have been described previously (DeBoer and Steffes, 1994). The objective of this laboratory system is to simulate the atmosphere of a given planet and measure the microwave properties of gases under these conditions. It consists of three sub-systems: (1) the planetary atmospheric simulator, (2) the microwave measurement sub-system and (3) the data collection and analysis sub-system. Figure 1 shows a block diagram of the laboratory configuration.

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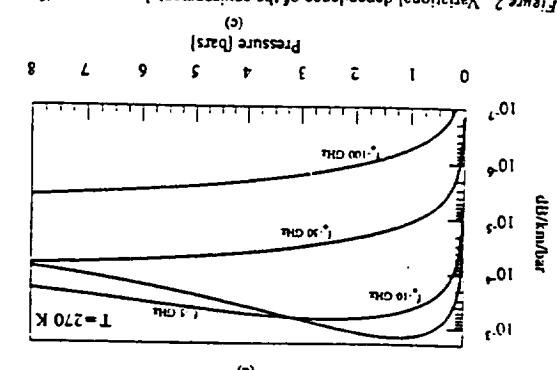
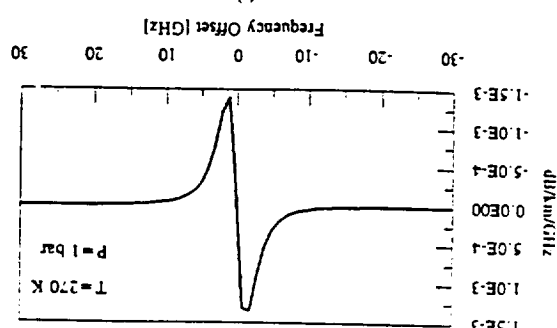
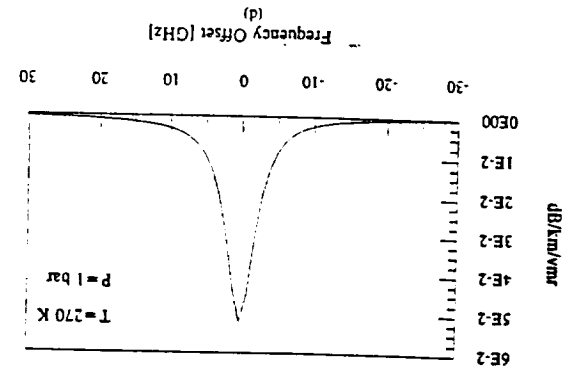
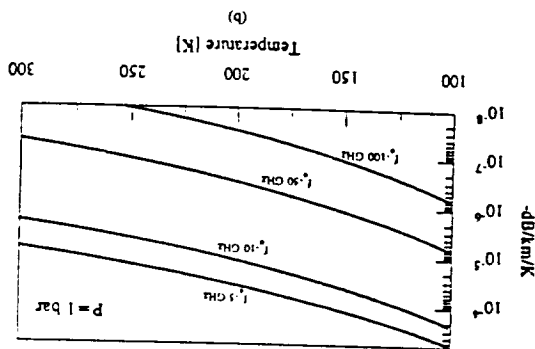


Figure 2. Variational dependence of the environmental parameters (frequency temperature, pressure, and mixing ratio) of one term of the mathematical formalism expression (Equation 4) for a 10% mixing ratio and typical line parameters of the 168 GHz H_2S line.

The planetary atmospheric simulator consists of a hermetically sealed stainless steel pressure vessel which in turn is placed inside a temperature chamber capable of reaching temperatures as low as 150 K or as high as 600 K. The pressure vessel is connected to three gas cylinders via stainless steel tubing with sufficient stop valves such that every piece of the network may be isolated to aid in the detection of leaks, as well as for safety considerations. A combustible gas detector and a water bubble technique were used to detect and locate leaks within the system. When properly sealed, the system can safely contain pressures exceeding 7 bars and sustain this pressure with no discernible leaks over a period of many hours. A roughing vacuum pump was also connected to the vessel to evacuate the chamber and is capable of producing a vacuum of 3 torr. For pressure measurements between 1 and 800 torr, a digital thermocouple vacuum gauge was used, while for higher pressures an analog gauge was used with an accuracy of 0.2 atm throughout its usable range.

The microwave sub-system consists of two cylindrical silver-plated cavity resonators which both reside within the pressure vessel. The larger of the two resonators has been used for S band (2.25 GHz or 13.3 cm) and X band (8.5 GHz or 3.5 cm) measurements which correspond to TE₀₁₃ and TE₀₆₇ modes respectively. The small resonator has been used for K band (21.7 GHz or 1.38 cm) measurements which corresponds to the TE₀₁₄ mode. Many other resonances are available for use at other wavelengths (see Figure 3). The three bands discussed above were used due to their relatively large quality factors. Mode suppression slots are incorporated in each resonator to block the degenerate TM modes which have lower quality factors as well as to let the gas flow freely into the resonators. Two HP (Hewlett-Packard) 8690 microwave sweep oscillators are used to generate the input signal in the three bands and a high resolution HP 8562B spectrum analyzer is used to measure the output signal. Initially a 10 dB attenuator was used on the output of the sweepers to provide 20 dB of return-loss isolation for the sweeper as it swept through the resonance. Later, a 40 dB return-loss ferrite isolator was used at S and X bands which greatly improved the isolation allowing the use of greater output power from the microwave sweeper as well as providing 10 dB extra throughout. This results in an approximate 15 dB increase in the signal-to-noise ratio for these bands. At K band, the 10 dB attenuator is still used.

The final sub-system (the data collection and analysis sub-system) consists of a personal computer and software package which reads the data in and allows data processing and resonance line-fitting to be performed (see appendix). This interface greatly improves the sensitivity and reliability of the experimental system from previous configurations, when the operator was required to eye-fit the half-power and center frequency points on the spectrum analyzer display. Full control of the spectrum analyzer is possible and batch file input gives speed and flexibility to this control. The following section describes in more detail this sub-system and other issues of the data processing.

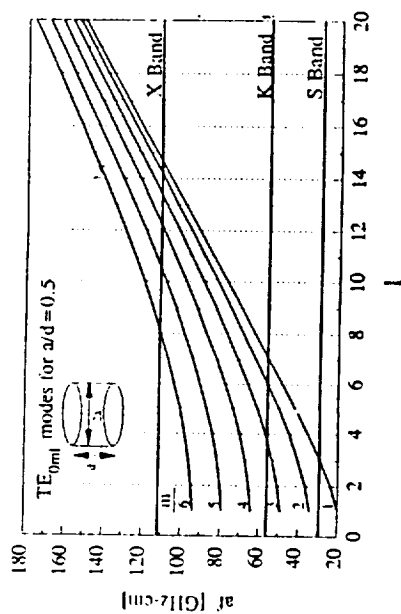


Figure 3. TE modes for the cylindrical resonators. Modes exist at the intersections of the curves and the vertical lines.

3. The Data Collection and Analysis Subsystem

As mentioned previously, in the past, data collection of the resonance parameters (i.e., bandwidth, center frequency and level) were eye-fit and read from the spectrum analyzer display. The disadvantages of this technique are manifold, not the least of which is operator fatigue near the end of a long series of data collection which degrades the quality of the results. The obvious solution is to read the data into a personal computer for analysis. A General Purpose Interface Bus (GPIB) was used to interface the spectrum analyzer and computer.

Data is read from the spectrum analyzer into a data buffer on the computer. A user-selectable Hamming window (typically a 50-point window) is convolved with the 600 data points read from the Hewlett-Packard 8562B spectrum analyzer to smooth the data and a simple peak searching routine then finds the center frequency and amplitude. The half power points are then found and the bandwidth computed. The spectrum, along with the smoothed fit, are displayed and the operator is queried as to whether this data point is to be used or not. (Occasionally, incomplete spectra are recorded which must be discarded.) If complete, the resonator measurement values are written to a data file. These values are:

- center frequency
- amplitude at the resonance center
- half power bandwidth
- quality factor
- an asymmetry figure of merit.

Date of run: 03/28/1994 Time of run: 14:40:03
 X band at vacuum (3 torr)
 3.95 mV, -60°C

<<Spectrum Analyzer Parameters>>				
Center frequency:	8541.81 MHz	span:	50.00 kHz	
RBW:	1.00 kHz	VBW:	1.00 kHz	
sweep time:	20.0000 sec	ref level:	-76.00 dBm	

Filter: 50 point Hanning

BW (kHz)	peak (MHz)	Q	level (dBm)	asymmetry (%)
71.190	8541.8032	116707	-76.94	5.03030
74.712	8541.8022	114330	-76.98	10.23102
74.965	8541.8032	113944	-77.01	0.63789
71.414	8541.8029	119609	-76.87	5.31724
71.697	8541.8007	115904	-76.87	12.37458
74.712	8541.8019	114330	-76.93	8.91089

6 data points

<BW (kHz)> = 73.7817 ± 1.34866
 <peak (MHz)> = 8541.8 ± 0.00190367
 <Q> = 115804 ± 2149.81
 <level (dBm)> = -76.9367 ± 0.0571544
 <asymmetry> = 7.12369 ± 4.22154
 <asymmetry> = 7.12369 ± 4.22154

Date of run: 03/28/1994 Time of run: 14:44:20

Figure 4. Sample LAB mode data file for X band (see appendix). Note the output format: Date and time stamp; Comments; Spectrum analyzer parameters; Filter used; Data points for particular lab sequence; and Mean and standard deviation of the data.

The asymmetry figure of merit, af , is defined as

$$af = 100 \frac{(f_r - f_c) - (f_c - f_l)}{f_r - f_l} \% \quad (7)$$

where f_r , f_l and f_c are the higher frequency half-power (-3 dB) point, lower frequency half-power (-3 dB) point and center of the resonance, respectively. It varies from $\pm 100\%$, with 0% being perfectly symmetrical. Typical asymmetry factors range between $\pm 10\%$. (Excessive asymmetry may suggest contamination of the data from the effects of adjacent resonances, and the results may be unreliable. If it persists for more than one scan, the system must be dismantled and thoroughly checked.) When all data have been taken and recorded, the program computes the averages and variance of all quantities, including the absolute value of the asymmetry as well as the asymmetry itself. These values are written to a data file and used in the computation of the absorptivity and its associated error. Typically,

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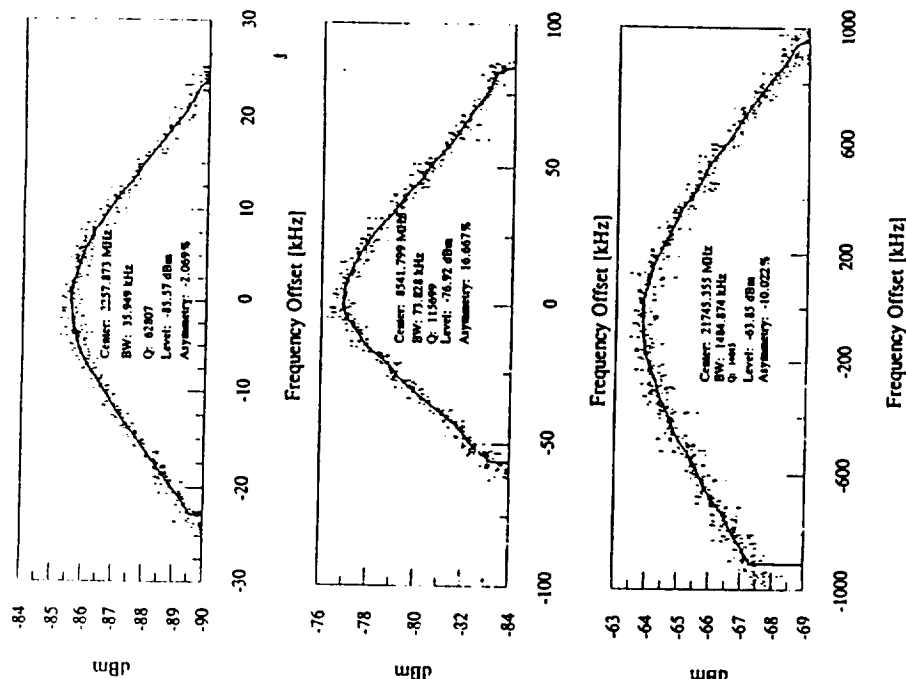


Figure 5. Representative spectra at the three bands used.

20 data points are taken per temperature, pressure and frequency to develop the error statistics.

The above sequence, beginning from reading a spectrum into the data buffer to computing the variances of a set of measurements and writing to an output data file, describes what is known as the "LAB mode" of the computer software PCSA, which is discussed in the appendix. Figure 4 shows the output data file for

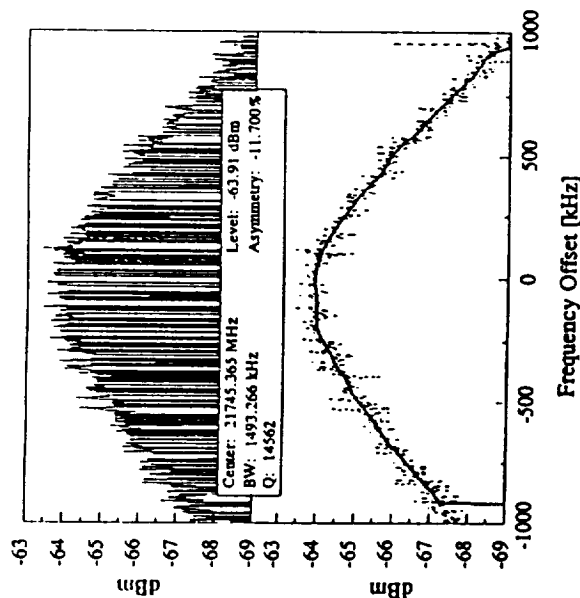


Figure 6. Same K-band spectrum as in the previously figure with artificially generated sweep-on-scans nulls along with the FIXed spectrum and new resonance fit.

six individual measurements at X band (8.5 GHz). Figure 5 shows representative spectra at the three bands, as well as the associated parameter fits for the smoothed data.

One problem encountered in fitting the spectrum is the presence of sweep-on-scans nulls. These occur when a sweeping source is used along with a sweeping bandpass filter on a spectrum analyzer so that at a given time energy from the signal may not be present in the given small frequency range received instantaneously by the spectrum analyzer. One solution is to slow the sweep time on the spectrum analyzer. In some cases, however, totally eliminating these nulls requires unacceptably long sweep times. Another is to utilize a feature of the data processing software called "FIX". FIX is a multi-pass averaging scheme whereby "null" data points are set to an average of its ten nearest neighbors until the change of all points to be FIXed is less than a one percent threshold.

Figure 6 is the same spectrum as the K-band spectrum in the previous figure with artificially generated sweep-on-scans nulls. The spectrum was then FIXed and new fits were computed. Note that the values of the original and manipulated data agree very well, as summarized below in percent differences:

- center frequency: 0.00005%

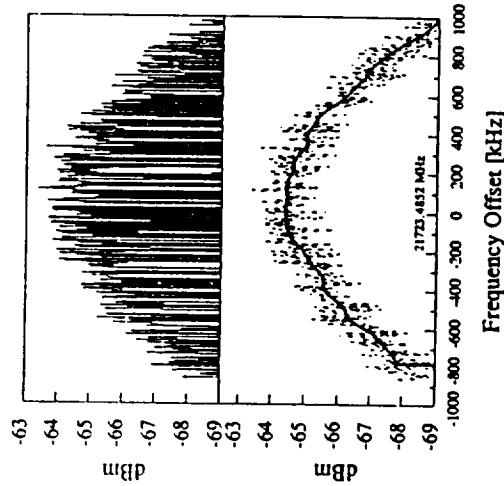


Figure 7. K-band spectrum with measured sweep-on-scans nulls along with the FIXed and fit spectra. The parameters fits are: $\Delta\nu = 1470.894$ kHz, $Q = 14769$, Level = -64.38 dBm and asymmetry = 8.42% .

Table I

System root mean variance at S, X and K bands

	Before	After
S band	0.061 dB/km	0.044 dB/km
X band	0.205 dB/km	0.143 dB/km
K band	1.293 dB/km	0.682 dB/km

- half-power bandwidth: 0.56%
- Q : 0.57%
- peak level: 0.09%
- asymmetry: 16.7%.

Figure 7 shows a K-band spectrum exhibiting measured sweep-on-scans nulls along with the FIXed and fit spectrum.

4. Conclusion

The computer interface and data processing software have greatly increased the sensitivity and reliability of the already highly sensitive microwave measurement

system at the Georgia Institute of Technology. Calculating the standard deviation for 10 measurements of the FWHM bandwidth before and after the inclusion of the computer interface shows marked improvement. For example, the standard deviation of 10 measurements of H₂S opacity at 6 bars in an H₂/He atmosphere at 213 K showed improvements of 34%, 41% and 16% at S, X and K bands respectively. It is probable that the improvement in reliability is greater than represented by this since an inherent viewer bias is present whenever eye-fit measurements are made on a noisy display. Another factor added by the computer-aided system is that more sample points can be easily taken to further increase the sensitivity. Table I shows the square root of the mean measurement variance of the system (the "standard deviation") at the three bands for all temperatures and pressures before and after the inclusion of the computer interface. For comparison, a resonator-based system similar to the K band resonator but operating between the frequencies 9.18 and 17.44 GHz (Spilker, 1990) had root mean variances of 1.02 and 2.46 dB/km, at frequencies corresponding to the two end-points.

Acknowledgements

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Appendix: The Computer Software - PCSA

The computer interface software consists of one stand-alone program called PCSA (Personal Computer-Spectrum Analyzer) written in the C programming language. PCSA is a case insensitive command-line driven system of which many commands have optional arguments, denoted below in square brackets []. Most commands can be abbreviated to the shortest possible number of unique characters. General PCSA commands are:

FILTER

DOS command sub-processor.

Set smoothing filter. Any filter can supplied by the user. Currently supported are 10, 20, 30, 40 and 50 point Hamming windows (out of 600 points comprising a spectrum from the IIP 8562).

FIT [SPECTRUM]

Fit the bandwidth etc. to a smoothed spectrum [take new spectrum]. If there is no data in the buffer, a new spectrum will automatically be taken.

FIX [PLOT]

FIX raw data buffer [and plot].

HELP?

On-line help facility.

LAB [nn]

Enter PLANETARY lab mode [and auto-take nn samples with no check]. LOAD 'file'

Read batch processing commands from 'file'.

PARAMETERS ['file']

Write spectrum analyzer parameters to screen/'file'.

PLOT

Plot spectrum in buffer.

QUERY 'xxx'?

Ask spectrum analyzer for data 'xxx'. E.g. 'query cf?' for center frequency data for the 8562.

QUIT, Q

Exit this program.

READ 'file'

Read 'file' into data buffer.

SAVE 'file'

Save buffer to 'file'.

SET FIX

Toggles between FIX being ON and OFF for lab mode.

SMOOTH [SAVE]

Smooth and plot spectrum in buffer, [and write to file (current directory)]. If there is no data in the buffer, a new spectrum will automatically be taken.

SPECTRUM [SAVE]

Write spectrum to raw data buffer and plot [and write to file (current directory)].

TRANSFORM [SAVE]

Take, view [and save (current directory)] the Fourier transform of the buffer.

All commands supported by the resident device.

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